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Statistical modeling of particles relative motion in a turbulent gas flow

I.V. Derevich *

Department of Thermodynamics and Heat Transfer, Moscow State University of Environmental Engineering, 21/4 Staraya Basmannaya Street, 107884 Moscow, Russia

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Abstract

On the base of modern probability approach the theoretical model of turbulent relative motion of particles in the turbulent flow is developed. Closed equation for probability density function of coordinates and velocities of two particles in turbulent flow is obtained. The system of equations for balance of mass, averaged velocities and intensities of turbulent chaotic motion of particles with account of correlated motion of particles are deduced. The closed expressions for intensity of relative chaotic motion between particles are obtained on the base of probability density function of particles displacement with correlation effects. The correlation functions, intensity of relative turbulent motion and relative diffusion coefficients of particles are numerically investigated. The calculation results are compared with data of large eddy simulations. The results of calculation intensity of droplets relative motion in atmospheric conditions are presented.

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1. Introduction

In gas flows the rate of particles or droplets coagulation depends on their relative velocity and collision frequencies. The relative velocity of particles is determined by the external forces, for example, mass forces as well as particles intensity of random motion in the turbulent flow. The paper is devoted to investigation the relative turbulent transport of particles with various sizes. The entrainment of particles in turbulence depends on their inertia. Small particles, whose dynamic relaxation time is much less than the integral time scale of turbulence are completely entrained in the turbulent motion of energy containing eddies. Without consideration the effect of particles inertia on the degree of entrainment in the small-scale turbulence, the averaged relative velocity between particles is determined by a gradient of a carrier phase velocity on a distance of order the sum of particles diameters [1,2]. In [3], within the framework of the model outlined in [1] the small-particles coagulation kernel

* Tel./fax: +7 095 362 5590. *E-mail address:* nchmt@iht.mpei.ac.ru

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was calculated with allowance for Brownian and relative turbulent diffusion. In [3] the effect of relative averaged velocity slips between particles due to gravity force was includes in efficiency of particles coagulation. In a gas flow small inertia particles have diameters lesser than Kolmogorov space micro scale. For such particles relative velocity due to gradient of fine grained turbulence on a distance of particles diameters is negligible. Trajectories of these particles are well correlated and chaotic relative velocity of small inertia particles equal to zero.

For inertial particles with dynamic relaxation time of order integral time macro scale of turbulence the intensity of their chaotic motion is determined by entrainment of particles into turbulent motion of energy containing eddies. These particles do not participate into small scale high frequency turbulence. In [4] it is assumed, that trajectories of inertial particles are not correlate. In [4] by analogy of kinetic theory of gaseous the energy of random motion of two particles was set as a sum of energy of chaotic motion of the particles. The degree of entrainment of particles into turbulent motion of large eddies was taken into account in [5]. In the [5] the approximate distribution

Nomenclature

$D_{\alpha,ik}$	coefficient of turbulent diffusion of ath particles	$\mathbf{W}_{\alpha\beta}$
$D_{\alpha\beta,ik}$	coefficient of turbulent relative diffusion be-	$\mathbf{w}_{\alpha\beta}$
	tween two particles	·
D_{\circ}	coefficient of turbulent diffusion of inertia less	Gree
	particles	γα
$d_{\rm p}$	diameter of a particle	Δ
$f_{\alpha}, f_{\alpha \beta}$	unconditional and conditional response func-	$\delta(\mathbf{x})$
	tions	3
$G_{\alpha}, G_{\alpha\beta}$	probability density functions of particles trans-	Λ
	fer	
g	gravity acceleration	λ
$\tilde{L}_{\rm E}$	Euler integral space scale	
N_{α}	distribution function of one type particles in	μ
	space	
$N_{\alpha\beta}$	distribution function of particles of two types in	$\rho_{\alpha\beta}$
•	space	σ_{α}, α
$q_{\alpha}, q_{\alpha \beta}$	unconditional and conditional response func-	
	tions	$ au_{lpha}$
Re_{λ}	Reynolds number calculated on Taylor micro-	$\Phi_{lphaeta}$
	scale	$\varphi_{\alpha\beta}$
$T_{\rm E}$	Euler integral temporary scale	
$T_{\rm L}$	Lagrange temporary scale	χ
T_{α}	temporary scale of gas velocity fluctuations	$\Psi_{\rm E}$
	along ath particle path	$\Psi^{(p)}_{\alpha}$
U	actual velocity of fluid phase	$\Psi^{(p)}$
u	velocity fluctuations of fluid phase	αιβ
$\mathbf{V}_{oldsymbol{lpha}}^{(\mathrm{p})}$	actual velocity of the ath particle	22α
\mathbf{V}_{α}	Euler velocity of ath particle	Sub
$\mathbf{v}^{(\mathrm{p})}_{\alpha}$	velocity fluctuations of α th particle	a B
$\mathbf{X}_{\alpha}^{(p)}$	Lagrange position of α th particle	α,p / \
xα	Euler position of ath particle	\ /
$Y_{\alpha\beta}, y_{\alpha\beta}$	relative distances between two particles	
Wα	averaged velocity of ath particle due to mass	
	force	

Greek symbols

-			
γα	nondimensional relative velocity of ath particle		
Δ	total dispersion of particles turbulent transfer		
$\delta(\mathbf{x})$	three-dimensional Dirac delta-function		
3	turbulent dissipation rate		
Λ	dispersion of particles turbulent transfer due to		
	inertia		
λ	dispersion of particles transfer with energy con-		
	taining eddies		
μ	ratio between Lagrange and Euler temporary		
	scales		
$\rho_{\alpha\beta}$	coefficient of two particles velocity correlation		
$\sigma_{\alpha}, \sigma_{\alpha\beta}$	second moments of particles velocity fluctua-		
	tions		
$ au_{lpha}$	dynamic relaxation time of α th particle		
$\Phi_{lphaeta}$	indicator function for two particles		
$\varphi_{\alpha\beta}$	probability density function of two particles		
	velocity distribution		
χ	structural parameter of turbulent flow		
$\Psi_{\rm E}$	Euler correlation function		
$arPsi_{oldsymbol{lpha}}^{(\mathrm{p})}$	unconditional gas velocity correlation function		
$\Psi^{(\mathrm{p})}_{\alpha \beta}$	conditional gas velocity correlation function		
Ω_{α}	parameter of inertia of α th particle		
Subscripts			
α,β	particles α th and β th types		
$\langle \rangle$	denotes result of averaging over an ensemble of		
\ /	turbulent realizations		

of turbulent energy of carrying phase was involved for calculation the intensity of random motion of particles with different sizes. But in [5] was assumed, that trajectories of inertial particles with equal sizes are completely correlated. So, in a turbulent gas flow inertial particles with equal diameters are not colliding with each other.

The large eddy simulations (LES) and direct numerical simulations was used in [6,7] for investigation the particles collisions in the homogeneous turbulent motion. In these works the role of trajectory correlations of inertial particles are brightly illustrated. In has been established that inertia less particles move in very correlated manner. With increasing particles inertia correlation between particles trajectories destroys and the relative turbulent velocity increases. For very inertial particles, whose dynamic relaxation times are mach larger then integral time scales of turbulence, the intensity of all turbulent motion of dispersed phase fall down. In [6] theoretical model for calculation the relative motion of particles with equal sizes was suggested. For calculation intensity of particles random motion was used Boltzmann hypothesis from kinetic theory of gaseous. The approach [6] is valid for particles with equal sizes and do not take into account effect of reduction of correlation between particles with increasing relative distance. In the models [6] the relative turbulent diffusion of particles is not considered. But the contribution of relative turbulent diffusion of particles in relative turbulent motion of particles is very important.

The perspective modern approach for investigation particles relative chaotic motion based on probability density function (PDF) for particles coordinates and velocities was suggested in [8]. The closing PDF equation has been achieved due to assumption that relative displacement of particles is a result only of relative random velocity between particles. The model [8] is valid for description relative chaotic motion of particles with equal sizes. We can note that in the specific case of particles with equal diameters without mass forces the results of present work coincides with data in [8].

In the present paper we developed a theoretical model of relative turbulent motion of particles with different diameters. We include the average relative velocities of particles due to mass forces. The closed PDF equation for coordinates and velocities of two different particles in inhomogeneous turbulence was obtained. Closed system of equations for describing processes of mass transfer due to particles relative motion is found. For closing terms, which describe correlation between particles trajectory, hypothesis by Corrsin [9] is involved. The calculation results are compared with LES data. Effects connected with particles inertia, various particles sizes and averaged velocities slip of particles is illustrated on an example of turbulent flow at atmospheric conditions.

2. Equation for PDF

2.1. Averaged and fluctuating quantities

In the present paper we do not investigate the variation the turbulent parameters of carrying gas due to presence of dispersed phase. In the gas flow the diameters of particles is lesser than Kolmogorov space micro scale. Equations for relative motion of two spherical solid particles α , β in a gas flow may be written in a form

$$\frac{\mathbf{d}\mathbf{V}_{\alpha}^{(\mathrm{p})}}{\mathbf{d}t} = \frac{1}{\tau_{\alpha}} \left(\mathbf{U} \left(\mathbf{X}_{\alpha}^{(\mathrm{p})}, t \right) + \mathbf{W}_{\alpha} - \mathbf{V}_{\alpha}^{(\mathrm{p})} \right), \quad \frac{\mathbf{d}\mathbf{X}_{\alpha}^{(\mathrm{p})}}{\mathbf{d}t} = \mathbf{V}_{\alpha}^{(\mathrm{p})}, \tag{1}$$

where $\mathbf{U}(\mathbf{x},t)$ is the velocity of gas phase; \mathbf{x} is Euler coordinate; $\mathbf{X}_{\alpha}^{(p)}(t)$, $\mathbf{V}_{\alpha}^{(p)}(t)$ is position and velocity of a α th particle; $\mathbf{W}_{\alpha} = \tau_{\alpha} \mathbf{g}$ is average relative particle velocity due to mass force; \mathbf{g} is acceleration due to mass force, for example, gravity; τ_{α} is particle relaxation time, which is dependent on particle relative velocity (see, for example, [10]).

Eq. (1) are present in the Lagrange variables. For passing from Lagrange variables to Euler variables in Eq. (1), we definite instantaneous two-point indicator function

$$\begin{split} \Phi_{\alpha\beta}(\mathbf{x}_{\alpha}, \mathbf{V}_{\alpha}, \mathbf{x}_{\beta}, \mathbf{V}_{\beta}, t) &= \delta(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(\mathrm{p})})\delta(\mathbf{V}_{\alpha} - \mathbf{V}_{\alpha}^{(\mathrm{p})}) \\ &\times \delta(\mathbf{x}_{\beta} - \mathbf{X}_{\beta}^{(\mathrm{p})})\delta(\mathbf{V}_{\beta} - \mathbf{V}_{\beta}^{(\mathrm{p})}), \end{split}$$
(2)

where $\mathbf{X}_{\alpha}^{(p)}, \mathbf{X}_{\beta}^{(p)}$ and $\mathbf{V}_{\alpha}^{(p)}, \mathbf{V}_{\beta}^{(p)}$ instantaneous positions and velocities of α th and β th particles, $\delta(\mathbf{x})$ is three dimensional Dirac delta-function.

After definition the function (2) we may speak about dispersed phase as a continuum fluid. Distribution of two particles in space and conditional velocity of α th particles are expressed through the indicator function (2)

$$N_{\alpha\beta}(\mathbf{x}_{\alpha}, \mathbf{x}_{\beta}, t) = \delta(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(p)})\delta(\mathbf{x}_{\beta} - \mathbf{X}_{\beta}^{(p)}),$$

$$= \int d\mathbf{V}_{\alpha} \int \boldsymbol{\varPhi}_{\alpha\beta} d\mathbf{V}_{\beta}, \qquad (3)$$

$$N_{\alpha\beta} \widetilde{\mathbf{V}}_{\alpha}(\mathbf{x}_{\alpha} | \mathbf{x}_{\beta}, t) = \mathbf{V}_{\alpha}^{(p)}(t)\delta(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(p)})\delta(\mathbf{x}_{\beta} - \mathbf{X}_{\beta}^{(p)})$$

$$= \int d\mathbf{V}_{\alpha} \int \mathbf{V}_{\alpha} \boldsymbol{\varPhi}_{\alpha\beta} d\mathbf{V}_{\beta}, \qquad (4)$$

where $\widetilde{\mathbf{V}}_{\alpha}(\mathbf{x}_{\alpha}|\mathbf{x}_{\beta}, t)$ is conditional velocity of α th particle in a point \mathbf{x}_{α} at the moment of time *t* provided that β th particle is located in the point \mathbf{x}_{β} at the same moment time.

Distribution of α th particle and unconditional velocity of dispersed phase of α th particles are follow from expressions (3) and (4)

$$N_{\alpha}(\mathbf{x}_{\alpha}, t) = \int d\mathbf{V}_{\alpha} \int d\mathbf{V}_{\beta} \int \Phi_{\alpha\beta} d\mathbf{x}_{\beta}$$
$$= \int N_{\alpha\beta}(\mathbf{x}_{\alpha}, \mathbf{x}_{\beta}, t) d\mathbf{x}_{\beta}, \qquad (5)$$

$$N_{\alpha}\mathbf{V}_{\alpha}(\mathbf{x}_{\alpha},t) = \mathbf{V}_{\alpha}^{(p)}(t)\delta(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(p)})$$
$$= \int d\mathbf{V}_{\alpha} \int d\mathbf{V}_{\beta} \int \mathbf{V}_{\alpha} \Phi_{\alpha\beta} d\mathbf{x}_{\beta}.$$
(6)

It is worth to note, that conditional and unconditional quantities are not equal

$$\mathbf{V}_{\alpha}(\mathbf{x}_{\alpha},t) \neq \mathbf{V}_{\alpha}(\mathbf{x}_{\alpha}|\mathbf{x}_{\beta},t).$$

After averaging over an ensemble of turbulent realization from definitions (2)–(6), we obtain averaged two-point PDF, two-point particle distribution, and averaged velocity of dispersed phase

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(...).

$$\begin{split} \left\langle \Phi_{\alpha\beta}(\mathbf{x}_{\alpha}, \mathbf{V}_{\alpha}, \mathbf{x}_{\beta}, \mathbf{V}_{\beta}, t) \right\rangle &= \delta \left\langle (\mathbf{x}_{\alpha} - \mathbf{R}_{\alpha}^{(p)}) \delta(\mathbf{V}_{\alpha} - \mathbf{V}_{\alpha}^{(p)}) \right\rangle \\ &\times \delta(\mathbf{x}_{\beta} - \mathbf{R}_{\beta}^{(p)}) \delta(\mathbf{V}_{\beta} - \mathbf{V}_{\beta}^{(p)}) \right\rangle, \\ \left\langle N_{\alpha\beta}(\mathbf{x}_{\alpha}, \mathbf{x}_{\beta}, t) \right\rangle &= \left\langle \delta(\mathbf{x}_{\alpha} - \mathbf{R}_{\alpha}^{(p)}) \delta(\mathbf{x}_{\beta} - \mathbf{R}_{\beta}^{(p)}) \right\rangle \\ &= \int d\mathbf{V}_{\alpha} \int \left\langle \Phi_{\alpha\beta} \right\rangle d\mathbf{V}_{\beta}, \tag{7} \\ \left\langle N_{\alpha\beta} \right\rangle \left\langle \widetilde{\mathbf{V}}_{\alpha}(\mathbf{x}_{\alpha} | \mathbf{x}_{\beta}, t) \right\rangle &= \left\langle \mathbf{V}_{\alpha}^{(p)}(t) \delta(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(p)}) \delta(\mathbf{x}_{\beta} - \mathbf{X}_{\beta}^{(p)}) \right\rangle \\ &= \int d\mathbf{V}_{\alpha} \int \mathbf{V}_{\alpha} \left\langle \Phi_{\alpha\beta} \right\rangle d\mathbf{V}_{\beta}. \tag{8} \end{split}$$

Instantaneous particle velocity we combine as the sum of the conditional averaged velocity at the points $\mathbf{x}_{\alpha} = \mathbf{X}_{\alpha}^{(p)}(t)$, $\mathbf{x}_{\beta} = \mathbf{X}_{\beta}^{(p)}(t)$ and fluctuating part

$$\begin{aligned} \mathbf{V}_{\alpha}^{(p)}(t) &= \left\langle \widetilde{\mathbf{V}}_{\alpha}(\mathbf{x}_{\alpha}|\mathbf{x}_{\beta},t) \right\rangle + \mathbf{v}_{\alpha}^{(p)}(t) \\ &= \left\langle \widetilde{\mathbf{V}}_{\alpha}(\mathbf{X}_{\alpha}^{(p)}(t)|\mathbf{X}_{\beta}^{(p)}(t),t) \right\rangle + \mathbf{v}_{\alpha}^{(p)}(t). \end{aligned}$$
(9)

We define the fluctuating component of the dispersed phase velocity with two-point distribution as

$$N_{\alpha\beta}\mathbf{v}_{\alpha} = \mathbf{v}_{\alpha}^{(\mathrm{p})}(t)\delta\left(\mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(\mathrm{p})}\right)\delta\left(\mathbf{x}_{\beta} - \mathbf{X}_{\beta}^{(\mathrm{p})}\right).$$
(10)

From relations (3), (4), (9), (10) are follows, that the value of correlation between two-point particle distribution and velocity fluctuation of the dispersed phase α th particles is zero

$$\langle N_{\alpha\beta}\mathbf{v}_{\alpha}\rangle = \left\langle \mathbf{v}_{\alpha}^{(\mathrm{p})}(t)\delta(\mathbf{x}_{\alpha}-\mathbf{X}_{\alpha}^{(\mathrm{p})})\delta(\mathbf{x}_{\beta}-\mathbf{X}_{\beta}^{(\mathrm{p})})\right\rangle = 0.$$

The previous definition of fluctuating part of particles velocity (10) corresponds well with the common definitions of the fluctuating component and PDF

$$\int d\mathbf{V}_{\alpha} \int (\mathbf{V}_{\alpha} - \langle \mathbf{V}_{\alpha} \rangle) \langle \boldsymbol{\varPhi}_{\alpha\beta} \rangle d\mathbf{V}_{\beta} = \int d\mathbf{V}_{\alpha} \int \mathbf{v}_{\alpha} \langle \boldsymbol{\varPhi}_{\alpha\beta} \rangle d\mathbf{V}_{\beta} = 0.$$

Unconditional particles space distribution and averaged velocity are following from expressions (5) and (6). The definition of averaged and fluctuating parts of dispersed phase was made by analogy as in previous paper [11].

2.2. Derivation of PDF equation

In the terms of fluctuating velocity of dispersed phase, by similar procedure, as in [11], we write down the equation for averaged PDF $\langle \Phi_{\alpha\beta}(\mathbf{x}_{\alpha},\mathbf{v}_{\alpha},\mathbf{x}_{\beta},\mathbf{v}_{\beta},t) \rangle$

$$\frac{\partial \langle \Phi_{\alpha\beta} \rangle}{\partial t} + (\langle V_{\alpha,k} \rangle + v_{\alpha,k}) \frac{\partial \langle \Phi_{\alpha\beta} \rangle}{x_{\alpha,k}} - \frac{\partial}{v_{\alpha,i}} \langle \Phi_{\alpha\beta} \rangle \left[\frac{\partial \langle V_{\alpha,i} \rangle}{\partial t} + (\langle V_{\alpha,k} \rangle + v_{\alpha,k}) \frac{\partial \langle V_{\alpha,i} \rangle}{\partial x_{\alpha,k}} - \frac{\langle U_{\alpha,i} \rangle + \tau_{\alpha} g_{i} - \langle V_{\alpha,i} \rangle}{\tau_{\alpha}} \right] - \frac{1}{\tau_{\alpha}} \frac{\partial}{\partial v_{\alpha,i}} v_{\alpha,i} \langle \Phi_{\alpha\beta} \rangle + (\langle V_{\beta,k} \rangle + v_{\beta,k}) \frac{\partial \langle \Phi_{\alpha\beta} \rangle}{x_{\beta,k}} - \frac{\partial}{v_{\beta,i}} \langle \Phi_{\alpha\beta} \rangle \left[\frac{\partial \langle V_{\beta,i} \rangle}{\partial t} + (\langle V_{\beta,k} \rangle + v_{\beta,k}) \frac{\partial \langle V_{\beta,i} \rangle}{\partial x_{\beta,k}} - \frac{\langle U_{\beta,i} \rangle + \tau_{\beta} g_{i} - \langle V_{\beta,i} \rangle}{\tau_{\beta}} \right] - \frac{1}{\tau_{\beta}} \frac{\partial}{\partial v_{\beta,i}} v_{\beta,i} \langle \Phi_{\alpha\beta} \rangle = \widehat{A} \langle \Phi_{\alpha\beta} \rangle,$$
(11)

$$\widehat{A}\langle \boldsymbol{\Phi}_{\alpha\beta} \rangle = -\frac{1}{\tau_{\alpha}} \frac{\partial}{\partial v_{\alpha,i}} \langle u_{i}(\mathbf{x}_{\alpha}, t) \boldsymbol{\Phi}_{\alpha\beta} \rangle - \frac{1}{\tau_{\beta}} \frac{\partial}{\partial v_{\beta,i}} \langle u_{i}(\mathbf{x}_{\beta}, t) \boldsymbol{\Phi}_{\alpha\beta} \rangle.$$
(12)

The operator $\widehat{A}\langle \Phi_{\alpha\beta}\rangle$ in the right hand side of Eq. (11) describes an interaction between turbulent energy containing eddies and particles. For obtaining the closed equation for PDF (11), it is necessary to find closed expression between turbulent velocity of continuous phase and PDF $\langle u_t(\mathbf{x}_{\alpha},t)\Phi_{\alpha\beta}\rangle$ and $\langle u_t(\mathbf{x}_{\beta},t)\Phi_{\alpha\beta}\rangle$.

We used the assumption about Gaussian approximation of the random velocity field of gas phase. With the assistance of the method of functional derivative (see, as example, [11]) we write down Furutsu–Novikov (Klyatskin [12]) expression for correlation between turbulent fluid velocity and PDF

$$\left\langle u_{i}(\mathbf{x}_{\alpha},t)\Phi_{\alpha\beta}\right\rangle = \int_{0}^{t} \mathrm{d}\xi \int \left\langle u_{i}(\mathbf{x}_{\alpha},t)u_{j}(\mathbf{y},\xi)\right\rangle \left\langle \frac{\delta\Phi_{\alpha\beta}}{\delta u_{j}(\mathbf{y},\xi)}\right\rangle \mathrm{d}\mathbf{y},$$
(13)

where $\langle \delta \Phi_{\alpha\beta} / \delta u_j(\mathbf{y}, \xi) \rangle$ is functional derivation of the twopoint PDF.

In expression (13) we take into account, that the main part of intensity of turbulent motion of particles is connected with energy containing eddies. In this assumption the functional derivation from PDF in (13) have the following form:

$$\frac{\delta \Phi_{\alpha\beta}}{\delta u_{j}(\mathbf{y},\xi)} = -\frac{\partial \Phi_{\alpha\beta}}{\partial x_{\alpha,k}} \frac{\delta x_{\alpha,k}^{(p)}(t)}{\delta u_{j}(\mathbf{y},\xi)} - \frac{\partial \Phi_{\alpha\beta}}{\partial v_{\alpha,k}} \frac{\delta v_{\alpha,k}^{(p)}(t)}{\delta u_{j}(\mathbf{y},\xi)} - \frac{\partial \Phi_{\alpha\beta}}{\partial x_{\beta,k}} \frac{\delta v_{\beta,k}^{(p)}(t)}{\delta u_{j}(\mathbf{y},\xi)} - \frac{\partial \Phi_{\alpha\beta}}{\partial v_{\beta,k}} \frac{\delta v_{\beta,k}^{(p)}(t)}{\delta u_{j}(\mathbf{y},\xi)}.$$
(14)

Expression (14) takes into account the involving particles in turbulent velocity fluctuations of carrier phase and random particles displacement due to turbulence. The fluctuating parts of particle velocity $v_{\alpha,k}^{(p)}$ and its displacement $x_{\alpha,k}^{(p)}$ we write down as

$$v_{\alpha,k}^{(\mathrm{p})}(t) = \frac{1}{\tau_{\alpha}} \int_{0}^{t} \exp\left(-\frac{t-s}{\tau_{\alpha}}\right) u_{k}(\mathbf{X}_{\alpha}^{(\mathrm{p})}(s), s) \,\mathrm{d}s,\tag{15}$$

$$x_{\alpha,k}^{(\mathrm{p})}(t) = \int_0^t \left[1 - \exp\left(-\frac{t-s}{\tau_\alpha}\right) \right] u_k(\mathbf{X}_\alpha^{(\mathrm{p})}(s), s) \,\mathrm{d}s. \tag{16}$$

Functional derivations from particle velocity and its displacement we calculate with assistance of expressions (15) and (16) (see, as example, [11])

$$\frac{\delta v_{\alpha,k}^{(p)}(t)}{\delta u_j(\mathbf{y},\xi)} = \frac{\delta_{jk}}{\tau_{\alpha}} \exp\left(-\frac{t-\xi}{\tau_{\alpha}}\right) \delta(\mathbf{y} - \mathbf{X}_{\alpha}^{(p)}(\xi)), \tag{17}$$

$$\frac{\delta x_{\alpha,k}^{(p)}(t)}{\delta u_{j}(\mathbf{y},\xi)} = \delta_{jk} \left[1 - \exp\left(-\frac{t-\xi}{\tau_{\alpha}}\right) \right] \delta(\mathbf{y} - \mathbf{X}_{\alpha}^{(p)}(\xi)), \quad (18)$$

where δ_{ik} is Kroneker delta.

As a result of substitution expressions (14), (17) and (18) into Eq. (13), we find expression for correlation $\langle u_i(\mathbf{x}_{\alpha},t)\Phi_{\alpha\beta}\rangle$

$$\langle u_{i}(\mathbf{x}_{\alpha},t)\Phi_{\alpha\beta}\rangle = -\langle u_{i}u_{j}\rangle f_{\alpha}\frac{\partial\langle \Phi_{\alpha\beta}\rangle}{\partial v_{\alpha,j}} - \langle u_{i}u_{j}\rangle\tau_{\alpha}q_{\alpha}\frac{\partial\langle \Phi_{\alpha\beta}\rangle}{\partial x_{\alpha,j}} - \langle u_{i}u_{j}\rangle f_{\beta|\alpha}\frac{\partial\langle \Phi_{\alpha\beta}\rangle}{\partial v_{\beta,j}} - \langle u_{i}u_{j}\rangle\tau_{\beta}q_{\beta|\alpha}\frac{\partial\langle \Phi_{\alpha\beta}\rangle}{\partial x_{\beta,j}}.$$

$$(19)$$

Here $\langle u_i u_j \rangle$ is second one-point moment of fluctuating velocity of gas phase, f_{α} , q_{α} , $f_{\beta|\alpha}$, $q_{\beta|\alpha}$ are unconditional and conditional response function of particles which describe entrainment of particles in turbulent fluctuation of the carrier phase

$$f_{\alpha} \langle u_{i} u_{j} \rangle = \frac{1}{\tau_{\alpha}} \int_{0}^{t} \exp\left(-\frac{t-\xi}{\tau_{\alpha}}\right) \langle u_{i}(\mathbf{x}_{\alpha}, t) u_{j}(\mathbf{X}_{\alpha}^{(p)}(\xi), \xi) \rangle d\xi, \quad (20)$$

$$q_{\alpha} \langle u_{i} u_{j} \rangle$$

$$= \int_{0}^{t} \left[1 - \exp\left(-\frac{t-\xi}{\tau_{\alpha}}\right) \right] \left\langle u_{i}(\mathbf{x}_{\alpha}, t) u_{j}\left(\mathbf{X}_{\alpha}^{(p)}(\xi), \xi\right) \right\rangle \mathrm{d}\xi,$$
(21)

$$f_{\beta|\alpha} \langle u_i u_j \rangle$$

$$= \frac{1}{\tau_{\beta}} \int_0^t \exp\left(-\frac{t-\zeta}{\tau_{\beta}}\right) \langle u_i(\mathbf{x}_{\alpha}, t) u_j\left(\mathbf{X}_{\beta}^{(p)}(\zeta), \zeta\right) \rangle d\zeta,$$
(22)

 $q_{\beta|\alpha} \langle u_i u_j \rangle = \int_0^t \left[1 - \exp\left(-\frac{t-\xi}{\tau_{\beta}}\right) \right] \langle u_i(\mathbf{x}_{\alpha}, t) u_j \left(\mathbf{X}_{\beta}^{(p)}(\xi), \xi\right) \rangle d\xi.$ (23)

Functions f_{α} , q_{α} in (20), (21) are unconditional; they depend only on the trajectory α th particle. Functions $f_{\beta|\alpha}$,

 $q_{\beta|\alpha}$ in (22), (23) are conditional, they depend on trajectory of β th particle, provided that particle α is located at the point \mathbf{x}_{α} at the moment of time *t*. From expressions (22) and (23) it can be expect, as the distance between particles increases, the conditional response function of particles decrease and the intensity of the relative motion of particles are determined only by the chaotic motion of the individual particles.

After substitution Eq. (19) in Eq. (12) we obtain closed form for expression describing the interaction of turbulence with two statistically connected particles

$$\begin{split} \widehat{A} \langle \Phi_{\alpha\beta} \rangle &= \langle u_{i}u_{j} \rangle \frac{f_{\alpha}}{\tau_{\alpha}} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\alpha,i} \partial v_{\alpha,j}} + \langle u_{i}u_{j} \rangle q_{\alpha} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\alpha,i} \partial x_{\alpha,j}} \\ &+ \langle u_{i}u_{j} \rangle \frac{f_{\beta}}{\tau_{\beta}} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\beta,i} \partial v_{\beta,j}} + \langle u_{i}u_{j} \rangle q_{\beta} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\beta,i} \partial x_{\beta,j}} \\ &+ \langle u_{i}u_{j} \rangle \left(\frac{f_{\beta|\alpha}}{\tau_{\alpha}} + \frac{f_{\alpha|\beta}}{\tau_{\beta}} \right) \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\alpha,i} \partial v_{\beta,j}} \\ &+ \langle u_{i}u_{j} \rangle \frac{\tau_{\beta}}{\tau_{\alpha}} q_{\beta|\alpha} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\alpha,i} \partial x_{\beta,j}} + \langle u_{i}u_{j} \rangle \frac{\tau_{\alpha}}{\tau_{\beta}} q_{\alpha|\beta} \frac{\partial^{2} \langle \Phi_{\alpha\beta} \rangle}{\partial v_{\beta,i} \partial x_{\alpha,j}}. \end{split}$$

$$(24)$$

Eq. (11) with expression (24) generates the closed form of PDF for two stochastically connected particles. The one-point PDF equation may be written down on the basis of the two-point equation for PDF (11), (24) as a result of integration over the variables \mathbf{x}_{β} , \mathbf{v}_{β} . This one-point equation is similar to the closed equation for PDF received earlier in [11].

2.3. Solution the PDF equation for uniform flow

In the case of steady uniform turbulent flow $\langle u_i u_j \rangle = \delta_{ij} \langle u_i^2 \rangle$ equation for PDF of particles velocities $\varphi_{\alpha\beta}(\mathbf{v}_{\alpha}, \mathbf{v}_{\beta})$ follows from (11), (12) and have the form

$$-\frac{1}{\tau_{\alpha}}\frac{\partial}{\partial v_{\alpha,i}}v_{\alpha,i}\langle\varphi_{\alpha\beta}\rangle - \frac{1}{\tau_{\beta}}\frac{\partial}{\partial v_{\beta,i}}v_{\beta,i}\langle\varphi_{\alpha\beta}\rangle$$
$$= \frac{\sigma_{\alpha,ii}^{\circ}}{\tau_{\alpha}}\frac{\partial^{2}\langle\varphi_{\alpha\beta}\rangle}{\partial v_{\alpha,i}\partial v_{\alpha,i}} + \frac{\sigma_{\beta,ii}^{\circ}}{\tau_{\beta}}\frac{\partial^{2}\langle\varphi_{\alpha\beta}\rangle}{\partial v_{\beta,i}\partial v_{\beta,i}} + \left(\frac{1}{\tau_{\alpha}} + \frac{1}{\tau_{\beta}}\right)\sigma_{\alpha\beta,ii}^{\circ}\frac{\partial^{2}\langle\varphi_{\alpha\beta}\rangle}{\partial v_{\alpha,i}\partial v_{\beta,i}}.$$
(25)

Here intensities of turbulent motion of particles in uniform approach are equal

$$\begin{aligned} \sigma_{\alpha,ii}^{\circ} &= \left\langle v_{\alpha,i}^{2} \right\rangle = \frac{f_{\alpha}}{\tau_{\alpha}} \left\langle u_{i}^{2} \right\rangle, \\ \sigma_{\beta,ii}^{\circ} &= \left\langle v_{\beta,i}^{2} \right\rangle = \frac{f_{\beta}}{\tau_{\beta}} \left\langle u_{i}^{2} \right\rangle, \\ \sigma_{\alpha\beta,ii}^{\circ} &= \left\langle v_{\alpha,i} v_{\beta,i} \right\rangle = \left(\frac{1}{\tau_{\alpha}} + \frac{1}{\tau_{\beta}}\right)^{-1} \left(\frac{f_{\beta|\alpha}}{\tau_{\alpha}} + \frac{f_{\alpha|\beta}}{\tau_{\beta}}\right) \left\langle u_{i}^{2} \right\rangle. \end{aligned}$$

$$(26)$$

Solution of the Eq. (25) has the form of Gaussian distribution for fluctuation velocities of two particles

$$\begin{split} \varphi_{\alpha\beta}(\mathbf{v}_{\alpha},\mathbf{v}_{\beta}) &= \prod_{i=1}^{3} \frac{1}{\sqrt{(1-\rho_{\alpha\beta}^{2})}\sigma_{\alpha,ii}^{\circ}\sigma_{\beta,ii}^{\circ}}} \\ &\times \exp\left[-\frac{1}{2(1-\rho_{\alpha\beta}^{2})} \left(\frac{v_{\alpha,ii}^{2}}{\sigma_{\alpha,ii}^{\circ}} + \frac{v_{\beta,ii}^{2}}{\sigma_{\beta,ii}^{\circ}} - \frac{2\rho_{\alpha\beta}v_{\alpha,i}v_{\beta,i}}{(\sigma_{\alpha,ii}^{\circ}\sigma_{\beta,ii}^{\circ})^{\frac{1}{2}}}\right)\right], \end{split}$$

$$(27)$$

where coefficient of correlation $\rho_{\alpha\beta}$ between velocity fluctuations of two particles is equal

$$ho_{lphaeta}=rac{\sigma^{\circ}_{lphaeta,ii}}{(\sigma^{\circ}_{lpha,ii}\sigma^{\circ}_{eta,ii})^{rac{1}{2}}}.$$

In uniform approach the expression for correlation coefficient between velocity fluctuations of two particles is

$$\rho_{\alpha\beta} = \frac{\tau_{\beta} f_{\beta|\alpha} + \tau_{\alpha} f_{\alpha|\beta}}{(\tau_{\alpha} + \tau_{\beta}) \sqrt{f_{\alpha} f_{\beta}}}.$$
(28)

From expression (27) one can find the probability density distribution of relative velocity $w_{\alpha\beta,i} = v_{\alpha,i} - v_{\beta,i}$ and mean velocity of two particles $v_{\alpha\beta,i} = (v_{\alpha,i} + v_{\beta,i})/2$

$$\begin{split} \varphi(\mathbf{v}_{\alpha\beta}) &= \int \varphi_{\alpha\beta}(\mathbf{v}_{\alpha\beta}, \mathbf{w}_{\alpha\beta}) \, \mathrm{d}\mathbf{w}_{\alpha\beta} \\ &= \prod_{i=1}^{3} \frac{1}{\sqrt{2\pi \left\langle v_{\alpha\beta,i}^{2} \right\rangle}} \exp\left(-\frac{v_{\alpha\beta,i}^{2}}{2 \left\langle v_{\alpha\beta,i}^{2} \right\rangle}\right), \\ \left\langle v_{\alpha\beta,i}^{2} \right\rangle &= \frac{1}{4} \left[\sigma_{\alpha,ii}^{\circ} + \sigma_{\beta,ii}^{\circ} + 2\rho_{\alpha\beta}(\sigma_{\alpha,ii}^{\circ}\sigma_{\beta,ii}^{\circ})^{\frac{1}{2}}\right], \\ \varphi(\mathbf{w}_{\alpha\beta}) &= \int \varphi(\mathbf{v}_{\alpha\beta}, \mathbf{w}_{\alpha\beta}) \, \mathrm{d}\mathbf{v}_{\alpha\beta} \\ &= \prod_{i=1}^{3} \frac{1}{\sqrt{2\pi \left\langle w_{\alpha\beta,i}^{2} \right\rangle}} \exp\left(-\frac{w_{\alpha\beta,i}^{2}}{2 \left\langle w_{\alpha\beta,i}^{2} \right\rangle}\right), \\ \left\langle w_{\alpha\beta,i}^{2} \right\rangle &= \left[\sigma_{\alpha,ii}^{\circ} + \sigma_{\beta,ii}^{\circ} - 2\rho_{\alpha\beta}(\sigma_{\alpha,ii}^{\circ}\sigma_{\beta,ii}^{\circ})^{\frac{1}{2}}\right]. \end{split}$$
(29)

Here $\langle v_{\alpha\beta,i}^2 \rangle$ and $\langle w_{\alpha\beta,i}^2 \rangle$ square of dispersions of turbulent fluctuations of mean and relative chaotic velocities of two particles.

In principle, the PDF $\langle \Phi_{\alpha\beta}(\mathbf{x}_{\alpha},\mathbf{v}_{\alpha},\mathbf{x}_{\beta},\mathbf{v}_{\beta},t)\rangle$ contains all information about hydrodynamics parameters of particles in inhomogeneous turbulent flow. However, it is very difficult to find the analytical or numerical solution of closed equation for PDF (11), (24) in a strongly inhomogeneous turbulence, and we are forced to turn to the system of first and second moments.

3. Equations for the first and second moments of particles velocity fluctuations

Out of the PDF Eqs. (11), (24), we can derive by the standard way the system for moments of dispersed phase velocity fluctuations.

The equations for space distribution of two particles and conditional averaged velocity have the form

$$\frac{\partial \langle N_{\alpha\beta} \rangle}{\partial t} + \frac{\partial \langle N_{\alpha\beta} \rangle \langle \widetilde{Y}_{\alpha,i} \rangle}{\partial x_{\alpha,i}} + \frac{\partial \langle N_{\alpha\beta} \rangle \langle \widetilde{Y}_{\beta,i} \rangle}{\partial x_{\beta,i}} = 0, \qquad (30)$$

$$\frac{\partial \left\langle \widetilde{V}_{\alpha,i} \right\rangle}{\partial t} + \left\langle \widetilde{V}_{\alpha,k} \right\rangle \frac{\partial \left\langle \widetilde{V}_{\alpha,i} \right\rangle}{\partial x_{\alpha,k}} + \frac{\partial \left\langle v_{\alpha,i} v_{\alpha,k} \right\rangle}{\partial x_{\alpha,k}} + \frac{\partial \left\langle v_{\alpha,i} v_{\beta,k} \right\rangle}{\partial x_{\beta,k}} \\
= \frac{\left\langle U_{\alpha,i} \right\rangle + \tau_{\alpha} g_{i} - \left\langle \widetilde{V}_{\alpha,i} \right\rangle}{\tau_{\alpha}} - \frac{D_{\alpha,ik}}{\tau_{\alpha}} \frac{\partial \ln \left\langle N_{\alpha\beta} \right\rangle}{\partial x_{\alpha,k}} \\
- \frac{1}{\tau_{\alpha}} \left(\tau_{\alpha} \left\langle v_{\alpha,i} v_{\beta,k} \right\rangle + \tau_{\beta} q_{\beta|\alpha} \left\langle u_{i} u_{k} \right\rangle \right) \frac{\partial \ln \left\langle N_{\alpha\beta} \right\rangle}{\partial x_{\beta,k}}, \quad (31)$$

 $D_{\alpha,ik} = \tau_{\alpha}(\langle v_{\alpha,i}v_{\alpha,j}\rangle + q_{\alpha}\langle u_iu_k\rangle),$

where $D_{\alpha,ik}$ is coefficient turbulent diffusion of α th particles.

From Eq. (31) one can see, that conditional averaged velocity of α th particles statistically depends on parameters of chaotic motion of β th particles.

The equation for second conditional moments of two particles velocity fluctuations also follows from Eqs. (11) and (24)

$$\frac{\partial \langle v_{\alpha,i}v_{\beta,j} \rangle}{\partial t} + \langle \widetilde{V}_{\alpha,k} \rangle \frac{\partial \langle v_{\alpha,i}v_{\beta,j} \rangle}{\partial x_{\alpha,k}} + \langle \widetilde{V}_{\beta,k} \rangle \frac{\partial \langle v_{\alpha,i}v_{\beta,j} \rangle}{\partial x_{\beta,k}} \\
+ \frac{1}{\langle N_{\alpha\beta} \rangle} \left(\frac{\partial \langle N_{\alpha\beta} \rangle \langle v_{\alpha,i}v_{\beta,j}v_{\alpha,k} \rangle}{\partial x_{\alpha,k}} + \frac{\partial \langle N_{\alpha\beta} \rangle \langle v_{\alpha,i}v_{\beta,j}v_{\beta,k} \rangle}{\partial x_{\beta,k}} \right) \\
+ \langle v_{\alpha,i}v_{\beta,k} \rangle \frac{\partial \langle \widetilde{V}_{\beta,j} \rangle}{\partial x_{\beta,k}} + \langle v_{\beta,j}v_{\alpha,k} \rangle \frac{\partial \langle \widetilde{V}_{\alpha,i} \rangle}{\partial x_{\alpha,k}} \\
= \left(\frac{1}{\tau_{\alpha}} + \frac{1}{\tau_{\beta}} \right) \left(\sigma_{\alpha\beta,ij}^{\circ} - \langle v_{\alpha,i}v_{\beta,j} \rangle \right), \\
\sigma_{\alpha\beta,ij}^{\circ} = \left(\frac{1}{\tau_{\alpha}} + \frac{1}{\tau_{\beta}} \right)^{-1} \left(\frac{f_{\beta|\alpha}}{\tau_{\alpha}} + \frac{f_{\alpha|\beta}}{\tau_{\beta}} \right) \langle u_{i}u_{j} \rangle,$$
(32)

where $\sigma^{\circ}_{\alpha\beta,ij}$ is correlation between velocity fluctuations of two particles in uniform approach.

Three first terms in the left hand side of Eq. (32) describe the nonstationary and convection effects due to averaged velocities of particles α and β types. Third moments in Eq. (32) present the turbulent transfer of second moments of particles velocity fluctuations. Terms with gradients from averaged particles velocities are generation the turbulent motion in dispersed phase. The term in the right hand side of Eq. (32) is source of particles turbulence due to involving of particles in motion of energy containing eddies.

From Eqs. (30)–(32) it can be seen, that space distribution of particles and conditional averaged velocity depend upon coordinates of both particles. The averaged parameters of a pair of particles depend on the mean $\mathbf{x}_{\alpha\beta} = (\mathbf{x}_{\alpha} + \mathbf{x}_{\beta})/2$ and relative $\mathbf{y}_{\alpha\beta} = \mathbf{x}_{\alpha} - \mathbf{x}_{\beta}$ coordinates. The scale of variation of the averaged parameters with respect to the relative coordinate $\mathbf{y}_{\alpha\beta}$ is substantially less than that with respect of the mean coordinate $\mathbf{x}_{\alpha\beta}$. From Eqs. (30)– (32) and defined relative variable $\mathbf{y}_{\alpha\beta}$ follow equation for relative distribution of particles in space and expression for relative averaged velocity of two particles

$$\frac{\partial \langle N_{\alpha\beta} \rangle}{\partial t} + \frac{\partial \langle N_{\alpha\beta} \rangle \langle W_{\alpha\beta,i} \rangle}{\partial y_{\alpha\beta,i}} = 0,$$

$$\langle \mathbf{W}_{\alpha\beta} \rangle = \left\langle \widetilde{\mathbf{V}}_{\alpha} \right\rangle - \left\langle \widetilde{\mathbf{V}}_{\beta} \right\rangle,$$

$$\langle W_{\alpha\beta,i} \rangle = (W_{\alpha,i} - W_{\beta,i}) - D_{\alpha\beta,ik} \frac{\partial \langle N_{\alpha\beta} \rangle}{\partial y_{\alpha\beta,k}}$$
(33)

$$+ (\tau_{\alpha} + \tau_{\beta}) \frac{\partial \langle v_{\alpha,i} v_{\beta,k} \rangle}{\partial y_{\alpha\beta,k}}, \qquad (34)$$

$$D_{\alpha\beta,ik} = D_{\alpha,ik} - \tau_{\alpha} (\langle v_{\alpha,i} v_{\beta,k} \rangle + q_{\alpha|\beta} \langle u_i u_k \rangle) + D_{\beta,ik} - \tau_{\beta} (\langle v_{\alpha,i} v_{\beta,k} \rangle + q_{\beta|\alpha} \langle u_i u_k \rangle).$$
(35)

Here, $D_{\alpha\beta,ik}$ is the coefficient of relative turbulent diffusion between two particles; $\mathbf{W}_{\alpha\beta}$ is averaged relative velocity between particles.

We note that with increasing in the distance between particles, movement of the particles becomes uncorrelated, and the relative diffusion coefficient $D_{\alpha\beta,ik}$ tends to the sum of the diffusion coefficients of the individual particles. The relative particle velocity is determined not only by the difference between the relative velocity due to the mass force (first term in the right hand side of Eq. (34)) but also by the gradients of the particles space conditional distribution (second term in the right hand side of Eq. (34)). Second term in the right hand side of Eq. (34) describes the relative diffusion of two particles. Last term in the right hand side of Eq. (34) depends on gradient of intensity of particles velocities correlation. This effect is analogous to the effect of turbophoresis in the case of individual particles in bounded flow [11]). From (33), (34) follow the equation for distribution of a pair of particles in space

$$\frac{\partial \langle N_{\alpha\beta} \rangle}{\partial t} + \frac{\partial}{\partial y_{\alpha\beta,i}} \left\{ \left[(W_{\alpha,i} - W_{\beta,i}) + (\tau_{\alpha} + \tau_{\beta}) \frac{\partial \langle v_{\alpha,i} v_{\beta,k} \rangle}{\partial y_{\alpha\beta,k}} \right] \langle N_{\alpha\beta} \rangle \right\}$$

$$= \frac{\partial}{\partial y_{\alpha\beta,i}} \left[D_{\alpha\beta,ik} \frac{\partial \langle N_{\alpha\beta} \rangle}{\partial y_{\alpha\beta,k}} \right].$$
(36)

Eq. (36) is similar to diffusion equation in the space of relative distances between two particles. Eq. (36) includes the relative velocities between particles due to mass force and relative diffusion between particles (term in right hand side (36)). The term in the left hand side of (36) with derivations from second moments of different particles velocity fluctuations can be interpret as additional turbophoretic force between particles. Explanation of this effect is following. Approaches of particles with each other increase the correlation between particles random velocities, so the gradient of second moments in (36) is negative. In [8] this turbophoretic force is used for explanation the preferential concentration of particles in isotropic turbulence.

The equation for second moment of two particles velocity fluctuation in the relative variables have the form

$$\frac{\partial \langle v_{\alpha,i} v_{\beta,j} \rangle}{\partial t} + \langle W_{\alpha\beta,k} \rangle \frac{\partial \langle v_{\alpha,i} v_{\beta,j} \rangle}{\partial y_{\alpha\beta,k}} + \frac{1}{\langle N_{\alpha\beta} \rangle} \frac{\partial \langle N_{\alpha\beta} \rangle \langle v_{\alpha,i} v_{\beta,j} w_{\alpha\beta,k} \rangle}{\partial y_{\alpha\beta,k}} \\
+ \langle v_{\alpha,i} v_{\beta,k} \rangle \frac{\partial \langle \widetilde{V}_{\beta,j} \rangle}{\partial x_{\alpha,k}} + \langle v_{\beta,j} v_{\alpha,k} \rangle \frac{\partial \langle \widetilde{V}_{\alpha,i} \rangle}{\partial x_{\beta,k}} \\
= \left(\frac{1}{\tau_{\alpha}} + \frac{1}{\tau_{\beta}}\right) (\sigma_{\alpha\beta,ij}^{\circ} - \langle v_{\alpha,i} v_{\beta,j} \rangle),$$
(37)

where $w_{\alpha\beta,k} = v_{\alpha,k} - v_{\beta,k}$ is fluctuation of relative velocity between particles.

From Eq. (37) it can be seen that the convective term with averaged relative velocity and the turbulent transport due to the relative fluctuating velocity of particles, as well as term associated with the generation of random motion of particles due to gradient of averaged dispersed phase velocity, contribute to the correlation of particles velocity fluctuations.

4. Calculation of particles response function

4.1. Unconditional response functions

Expressions (20) and (21) depend upon the gas velocity correlation along the trajectory of individual particle. At the moment of time t the α th particle should pass trough the point \mathbf{x}_{α} . Along a trajectory of the α th particle reasonably representation is valid

$$\mathbf{x}_{\alpha} = \mathbf{X}_{\alpha}^{(\mathrm{p})}(t) = \mathbf{X}_{\alpha}^{(\mathrm{p})}(t-\xi) + \mathbf{X}_{\alpha}^{(\mathrm{p})}(\xi).$$

The expression for fluctuation of velocity of carrier phase along the particle trajectory have a following from above formula

$$\left\langle u_{i}(\mathbf{x}_{\alpha},t)u_{j}\left(\mathbf{X}_{\alpha}^{(\mathrm{p})}(\xi),\xi\right)\right\rangle = \left\langle u_{i}(\mathbf{x}_{\alpha},t)u_{j}\left(\mathbf{x}_{\alpha}-\mathbf{X}_{\alpha}^{(\mathrm{p})}(t-\xi),\xi\right)\right\rangle,$$
(38)

In the approximation of local homogeneous and stationary turbulence the following representation for two-points and two-times correlation function of gas velocity fluctuation is fair

$$\left\langle u_i(\mathbf{x}_1, t_1)u_j(\mathbf{x}_2, t_2)\right\rangle = \left\langle u_iu_j\right\rangle \Psi_{\mathrm{E}}(\mathbf{x}_1 - \mathbf{x}_2, |t_1 - t_2|),\tag{39}$$

where $\Psi_{\rm E}(\mathbf{x},t)$ is Euler correlation function in the coordinate frame moving with the carrying phase averaged velocity.

With the assistance of expressions (38) and (39) we obtain the representation for fluid velocity correlation function along the particle trajectory $\Psi_{\alpha}^{(p)}(s)$

$$\langle u_{i}(\mathbf{x}_{\alpha},t)u_{j}(\mathbf{X}_{\alpha}^{(\mathrm{p})}(\xi),\xi)\rangle = \langle u_{i}u_{j}\rangle\langle \Psi_{\mathrm{E}}(\mathbf{X}_{\alpha}^{(\mathrm{p})}(t) - \mathbf{X}_{\alpha}^{(\mathrm{p})}(\xi),t-\xi)\rangle$$

$$= \langle u_{i}u_{j}\rangle\int \langle\delta(\mathbf{y}_{\alpha} - \mathbf{X}_{\alpha}^{(\mathrm{p})}(s))\Psi_{\mathrm{E}}(\mathbf{y}_{\alpha},s)\rangle d\mathbf{y}_{\alpha}$$

$$= \langle u_{i}u_{j}\rangle\Psi_{\alpha}^{(\mathrm{p})}(s),$$

$$(40)$$

where $\mathbf{X}_{\alpha}^{(p)}(s)$ is relative distance of particle transfer in the coordinate frame fixed with gas averaged velocity $\langle \mathbf{U} \rangle$.

In the course of independent averaging hypothesis by Corrsin [9,10] we write the expression for correlation of gas velocity fluctuations along the random trajectory of particle in (40)

$$\Psi_{\alpha}^{(p)}(s) = \left\langle u_{i}u_{j}\right\rangle \int \left\langle G_{\alpha}(\mathbf{y}_{\alpha},s)\right\rangle \Psi_{\mathrm{E}}(\mathbf{y}_{\alpha},s)\,\mathrm{d}\mathbf{y}_{\alpha},\tag{41}$$

where $\langle G_{\alpha}(\mathbf{y}_{\alpha},s)\rangle = \langle \delta(\mathbf{y}_{\alpha} - \mathbf{X}_{\alpha}^{(p)}(s))\rangle$ is probability density function of particle transfer to the distance \mathbf{y}_{α} during interval of time $s = t - \xi$.

The formula (41) mean, that gas velocity correlation $\Psi_{\alpha}^{(p)}(s)$ includes all particle trajectories, which reach the space point \mathbf{x}_{α} during interval of time $s = t - \xi$. Averaging in the right hand side of (41) is executed over the ensemble of random particle trajectories and over the ensemble of gas turbulent velocity fluctuations. The procedure of the function $\langle G_{\alpha}(\mathbf{y}_{\alpha},s) \rangle$ calculation is presented in Appendix A.

The expression for response functions (20) and (21) for the α th particle may be written as

$$f_{\alpha} = \frac{1}{\tau_{\alpha}} \int_{0}^{t} \exp\left(-\frac{s}{\tau_{\alpha}}\right) \Psi_{\alpha}^{(p)}(s) \,\mathrm{d}s,$$

$$q_{\alpha} = \int_{0}^{t} \left[1 - \exp\left(-\frac{s}{\tau_{\alpha}}\right)\right] \Psi_{\alpha}^{(p)}(s) \,\mathrm{d}s.$$
(42)

In account of expressions (42) the formula for coefficient of turbulent diffusion of particles in Eq. (31) becomes

$$D_{\alpha,ii} = \left\langle u_i^2 \right\rangle \int_0^t \Psi_{\alpha}^{(p)}(s) \,\mathrm{d}s. \tag{43}$$

From (43) we conclude that coefficient of diffusion of particles is function of gas velocity correlation along particle trajectory.

4.2. Conditional response functions

The main idea used at calculation of conditional response functions (22) and (23) we shall explain on an example of function $f_{\beta|\alpha}$

$$f_{\beta|\alpha}\langle u_i u_j \rangle = \frac{1}{\tau_{\beta}} \int_0^t \exp\left(-\frac{t-\zeta}{\tau_{\beta}}\right) \left\langle u_i(\mathbf{x}_{\alpha},t) u_j\left(\mathbf{X}_{\beta}^{(p)}(\zeta),\zeta\right) \right\rangle \mathrm{d}\zeta,$$

In the last expression conditional gas velocity correlation depends on a random trajectory of β th particle provided that α th particle at the moment of time *t* occupies position \mathbf{x}_{α} . Relative displacement of β th particle can be presented as

$$\mathbf{x}_{\alpha} - \mathbf{X}_{\beta}^{(p)}(\xi) = \mathbf{x}_{\alpha} - \mathbf{X}_{\alpha}^{(p)}(\xi) + \mathbf{Y}_{\alpha\beta}^{(p)}(\xi),$$

where $\mathbf{Y}_{\alpha\beta}^{(p)}(\xi) = \mathbf{X}_{\alpha}^{(p)}(\xi) - \mathbf{X}_{\beta}^{(p)}(\xi)$ is instantaneous relative distance between particles at the moment of time ξ .

Conditional gas velocity correlation along a trajectory of β th particle can be written as

$$\begin{split} \left\langle u_{i}(\mathbf{x}_{\alpha},t)u_{j}\left(\mathbf{X}_{\beta}^{(p)}(\xi),\xi\right)\right\rangle \\ &=\left\langle u_{i}u_{j}\right\rangle\int d\mathbf{y}_{\alpha}\int \left\langle \delta\left(\mathbf{y}_{\alpha}-\mathbf{X}_{\alpha}^{(p)}(s)\right)\right. \\ &\times \delta\left(\mathbf{Y}_{\alpha\beta}-\mathbf{Y}_{\alpha\beta}^{(p)}(\xi)\right)\Psi_{\mathrm{E}}\left(\mathbf{y}_{\alpha}+\mathbf{Y}_{\alpha\beta},s\right)\right\rangle d\mathbf{Y}_{\alpha\beta} \\ &=\left\langle u_{i}u_{j}\right\rangle\Psi_{\beta|\alpha}^{(p)}(s,\mathbf{y}_{\alpha\beta}), \end{split}$$
(44)

where $s = t - \xi$ and $\mathbf{y}_{\alpha\beta} = \mathbf{x}_{\alpha} - \mathbf{x}_{\beta}$ is relative distance between particles at the moment of time *t*.

The expression (44) is written in the coordinate frame moving with averaged gas velocity. Conditional correlation of carrier phase velocity fluctuation in (44) $\Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta})$ is a result of averaging over the ensemble of random realizations of two particle trajectories and over the ensemble of turbulence realizations. In the sense of suggestion by Corrsin [9,10] we rewrite (44) in the following form:

$$\begin{split} \Psi_{\beta|\alpha}^{(p)}(s,\mathbf{y}_{\alpha\beta}) &= \int \mathrm{d}\mathbf{y}_{\alpha} \int \langle G_{\alpha}(\mathbf{y}_{\alpha},s) \rangle \big\langle G_{\alpha\beta}(\mathbf{y}_{\alpha\beta},t|\mathbf{Y}_{\alpha\beta},\xi) \big\rangle \\ &\times \Psi_{\mathrm{E}}(\mathbf{y}_{\alpha}+\mathbf{Y}_{\alpha\beta},s) \,\mathrm{d}\mathbf{Y}_{\alpha\beta}, \end{split}$$
(45)

where $\mathbf{Y}_{\alpha\beta}$ is relative distance between particles at the current time ξ ; $\langle G(\mathbf{y}_{\alpha\beta}t|\mathbf{Y}_{\alpha\beta},\xi)\rangle$ is conditional probability density function for relative displacement of particles.

The expression (45) means, that the conditional gas velocity correlation $\Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta})$ includes contribution of two types of random trajectories. Fist, the contribution of α th particle random trajectories, which reach the space point \mathbf{x}_{α} at the moment of time *t*. And second, relative trajectories between α th and β th particles provided, that relative distance between two particles will be $\mathbf{y}_{\alpha\beta} = \mathbf{x}_{\alpha} - \mathbf{x}_{\beta}$ at the moment of time *t*.

In (45) function $\langle G(\mathbf{y}_{\alpha\beta}t|\mathbf{Y}_{\alpha\beta},\xi)\rangle$ takes into account all trajectories of two particles during time $t \ge \xi \gg \tau_{\alpha}, \tau_{\beta}$ which will be separated on the distance $\mathbf{y}_{\alpha\beta}$ at the moment of time *t*. Calculation of function $\langle G(\mathbf{y}_{\alpha\beta}t|\mathbf{Y}_{\alpha\beta},\xi)\rangle$ is described in Appendix A.

In the sense of conditional gas velocity correlation function $\Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta})$ the conditional response functions $f_{\beta|\alpha}$ and $q_{\beta|\alpha}$ in (22), (23) are depends on relative distance $\mathbf{y}_{\alpha\beta}$, and have the following form:

$$f_{\beta|\alpha}(\mathbf{y}_{\alpha\beta}) = \frac{1}{\tau_{\beta}} \int_{0}^{t} \exp\left(-\frac{s}{\tau_{\beta}}\right) \Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta}) \, \mathrm{d}s,$$

$$q_{\beta|\alpha}(\mathbf{y}_{\alpha\beta}) = \frac{1}{\tau_{\beta}} \int_{0}^{t} \left[1 - \exp\left(-\frac{s}{\tau_{\beta}}\right)\right] \Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta}) \, \mathrm{d}s.$$
(46)

For inertial particles $\tau_{\alpha} \approx \tau_{\beta} \gg T_{\rm E}$, as we can see from (A.16) and (A.18), the function $\langle G_{\alpha\beta}(\mathbf{y}_{\alpha\beta},t|\mathbf{Y}_{\alpha\beta},\xi)\rangle$ is decrease as $\langle G_{\alpha\beta}\rangle \propto \sqrt{T_{\rm E}/\tau_{\alpha}}$. In that case response function $f_{\beta|\alpha}$ in (46) reduced as $f_{\beta|\alpha} \propto (T_{\rm E}/\tau_{\alpha})^{3/2}$, and coefficient of particles velocity correlation (28) $\rho_{\alpha\beta} \rightarrow 0$. This result reflects the fact, that chaotic motion of inertial particles is uncorrelated. Increasing the relative distance $\mathbf{y}_{\alpha\beta}$ between particles leads to decreasing correlation between particles and causes reduction of conditional response functions (46).

Expression for coefficient of relative turbulent diffusion of two particles follows from (35) and (46)

$$D_{\alpha\beta,ii}(\mathbf{y}_{\alpha\beta}) = \int_0^t \left[\Psi_{\alpha}^{(p)}(s) - \Psi_{\alpha|\beta}^{(p)}(s, \mathbf{y}_{\alpha\beta}) \right] \mathrm{d}s + \int_0^t \left[\Psi_{\beta}^{(p)}(s) - \Psi_{\beta|\alpha}^{(p)}(s, \mathbf{y}_{\alpha\beta}) \right] \mathrm{d}s.$$
(47)

Coefficient of relative diffusion of two particles (47) depends on relative distance $y_{\alpha\beta}$. Turbulent motions of inertia less particles on a small distance $y_{\alpha\beta}$ are well correlated and coefficient of relative diffusion tends to zero. Increasing the distance $y_{\alpha\beta}$ destroys correlation between turbulent motion of the particles and relative diffusion coefficient increases.

5. Approximation of gas velocity correlations along particles trajectories

The Euler correlation function in Eq. (39) we approximate in the form, which is corresponds to energy containing eddies

$$\Psi_{\rm E}(y,s) = \exp\left(-\frac{y}{L_{\rm E}} - \frac{s}{T_{\rm E}}\right). \tag{48}$$

After substitution (48) into expression (41) we write down expression for unconditional gas velocity correlation along a particle trajectory

$$\Psi_{\alpha}^{(p)}(s) = e^{-\frac{s}{T_E}} \psi(L_E, \Delta_{\alpha}, W_{\alpha}).$$
(49)

In (49) the dispersion Δ_{α} is function of time *s* (see Appendix A), and for s = 0 the value $\Psi_{\alpha}^{(p)}(s) = 1$. Function $\psi(L_{\rm E}, \Delta_{\alpha}, W_{\alpha})$ in (49) is definite in Appendix B and has an enough complex structure that complicates calculations. Below we shall suggest simple formula for correlation (49).

For convenience further statement we define dimensionless parameters. Parameter of particle inertia we will calculate as ratio between particles dynamic relaxation time and Euler integral time scale $\Omega_{\alpha} = \tau_{\alpha}/T_{\rm E}$. In some publications (for example, [7,8]) the parameter of particles inertia Ω_{α} is denotes as Stokes number of particles.

Crossing trajectory effect we will account with the help of parameter $\gamma_{\alpha} = W_{\alpha}/u$ [14]. Turbulent temporary scales in Lagrange and Euler variables are different (see, for example, [10,13]). And we define the ratio between Lagrange and Euler integral time scales as $\mu = T_{\rm L}/T_{\rm E}$ ($T_{\rm L}$ is Lagrange time scale calculated along the trajectory of inertia less fluid particle). The structural parameter of turbulence, which is depends on flow types (see, for example, [10]) we denote as $\chi = uT_{\rm E}/L_{\rm E}$. Nondimensional relative mean square turbulent velocity between particle we denote as $w_{\alpha\beta}^{\circ} = w_{\alpha\beta}/u$. Nondimensional time we denote as $s^{\circ} = s/T_{\rm E}$ and $Y_{\alpha\beta}^{\circ} = y_{\alpha\beta}/L_{\rm E}$ is nondimensional distance between particles.

We calculate the integral time scale of fluid velocity correlations along a particle path

$$T_{\alpha} = \int_0^{\infty} \Psi_{\alpha}^{(p)}(s) \,\mathrm{d}s. \tag{50}$$

For particles without inertia $\Omega_{\alpha} \to 0$ and average velocity slip $\gamma_{\alpha} \to 0$ we calculate the ratio between Lagrange and Euler integral time scales $\mu \approx 0.6$. The calculation was made with account Eqs. (49), (A.8)–(A.11) in the assumption $\chi = 1$. For inertial particles with velocity slip function (49) and time scale (50) are depend on parameters Ω_{α} and γ_{α} . For inertial particles $\Omega_{\alpha} \gg 1$ correlation function $\Psi_{\alpha}^{(p)}(s)$ tends to Euler correlation (48) (see Appendix A) and integral time scale (50) aspires to $T_{\rm E}$.

We suggest simple exponential approximation of gas velocity correlation $\Psi_{\alpha}^{(p)}(s)$ along a particle trajectory

$$\Psi_{\alpha}^{(p)}(s) = \exp\left(-\frac{s}{T_{\alpha}}\right).$$
(51)



Fig. 1. Correlation functions: (1) Euler correlation; (2) Lagrange correlation; (3)–(5) correlation functions along a particle path. Solid lines are exact expression (49), dash lines are exponential approximation (51).



Fig. 2. Dependence the response functions (44) on particles inertia for various nondimensional velocities slip.

In Fig. 1 are shown correlation functions in exact presentation (49) and exponential approximation (51). One can see that Lagrange correlation function decreases rapidly than Euler correlation function. The increasing particle average velocity slip leads to decreasing the value of velocity correlation function along particle trajectory, which reflect decreasing contact time between particle and turbulent energy containing eddies. The consent of two ways of correlation representations is satisfactory.

Fig. 2 illustrates influence of particle inertia and average velocity slip on particle response function (44). The parameter of particle inertia Ω_{α} and parameters of crossing trajectory effect γ_{α} diminish the intensity of particle turbulent motion.

6. Calculation results

In nondimensional variables correlation coefficient between particles velocities (28) have the following form:

$$\rho_{\alpha\beta} = \frac{\Omega_{\beta} f_{\beta|\alpha} + \Omega_{\alpha} f_{\alpha|\beta}}{(\Omega_{\alpha} + \Omega_{\beta}) \sqrt{f_{\alpha} f_{\beta}}}.$$
(52)

For calculation the value of particles correlation coefficient (52) was build iteration procedure with numerical integration of expressions (44), (49) and numerical integration of expressions (47), (49), where was used (A.20) and (A.21). The behavior of correlation coefficient as a function of nondimensional relative distance between two identical particles is shown in Fig. 3. The correlation between particles motion monotonically decrease with increasing relative distance. For small distance between particles correlation coefficient is reduced with particles inertia. Correlation coefficient of inertial particles is more pronounced on larger distance $Y^{\circ}_{\alpha\beta}$ than for particles with lesser inertia. Fig. 4 presents correlation coefficients between two particles as a function of particles inertia. One can see, that turbulent motion of particles with small inertia $\Omega_{\alpha}^{-1} \gg 1$ are well correlated $\rho_{\alpha\beta} \rightarrow 1$. For particles with sufficient inertia



Fig. 3. Correlation coefficient of two identical particles with various inertia and velocity slip as a function of relative distance between particles.



Fig. 4. Influence parameter of inertia of two identical particles on correlation coefficient. Points are LES results [6].

 $\Omega_{\alpha}^{-1} \gg 1$ correlation between particles motion is destroyed. Correlation between particles with small inertia falls rapidly with increasing relative distance $Y_{\alpha\beta}^{\circ}$. For inertial particles correlation is preserve on sufficiently larger distances. The results of our calculations are well coordinated with LES data [6].

Tendencies in behavior of correlation of two particles are reflect the dependence of mean square turbulent relative velocity between particles (Fig. 5). For well correlated motion of small inertia particles $\Omega_{\alpha}^{-1} \gg 1$ turbulent relative velocity monotonically tends to zero. For very inertial particles $\Omega_{\alpha}^{-1} \gg 1$ the relative turbulent velocity also decreases. This tendency one can explain as a result of decreasing the entrainment of inertial particles in turbulence. So, we san see the maximum value of turbulent relative velocity in the diapason of particles inertia $\Omega_{\alpha} \approx 1$. Relative distance between small inertia particles rapidly destroys correlation in particles motion, which leads to increasing the relative velocity. Turbulent motion of inertial particles is correlated along the larger distances between particles, and chaotic relative velocities between particles is less depends on $Y^{\circ}_{\alpha\beta}$. Fig. 6 illustrates influence of particles velocity slip



Fig. 5. Dependence of two identical particles turbulent relative velocity on particles inertia. Points are LES results [6].



Fig. 6. Influence of average particles velocity slip on turbulent relative velocity between particles.

on turbulent relative velocities between particles. The increasing of velocity slip diminishes entrainment of particles into turbulence and decrease intensity of relative turbulent velocity. Data of LES [6] on Figs. 5 and 6 confirm results of our calculations.

Coefficient of particle diffusion (43) is calculated with the help of numerical integration the expression (49). Results of our calculations are shown on Fig. 7. On the figure we present the diffusion coefficient in homogeneous approach D_{α} , so we dropped indexes of coordinates. The diffusion coefficient monotonically decreases with increasing average particles velocity slip. This crossing trajectory effect is accounted in the well known Csanady approximation [15]. The diffusion coefficient of inertial particles with small velocity slip is larger than diffusion coefficient of inertia less particles. This tendency is explained as a result of aspiration integral time scale along trajectory of inertial particles (50) to Euler time scale $T_{\rm E} > T_{\rm L}$.

The behavior of relative diffusion of two identical particles illustrates Figs. 8 and 9. On the Fig. 8 is shown influence



Fig. 7. Particle diffusion coefficients as a function of nondimensional velocity slip. Dashed line is Csanady approximation [15].



Fig. 8. Dependence of relative diffusion coefficients between two identical particles on particles inertia. Dashed lines are usual turbulent diffusion coefficient of particles.



Fig. 9. Dependence of coefficient of relative diffusion on a distance between particles. Dashed lines are usual coefficient of particles diffusion.

of particles inertia on relative diffusion. One can see, that for small inertia particles there no relative turbulent diffusion. Increasing particles inertia leads to increasing relative diffusion, which can be explained as a result of decreasing the correlation motion between inertial particles. Value of relative diffusion coefficient is decreases as a function of averaged velocity slip, which is connected with crossing trajectory effect. During distraction a turbulent correlation between particles the coefficient of relative diffusion aspires to the sum of turbulent diffusion coefficients D_{α} . Tendency of increasing the relative turbulent diffusion between particles we can see on the Fig. 9. Increasing relative distance between particles leads to increasing coefficient of particles relative turbulent diffusion. For small inertia particles the growth of the relative diffusion is more rapidly then with larger inertia. Particles velocity slip diminishes both coefficients of turbulent diffusion.

We calculate the mean square turbulent relative velocities between water droplets in the typical atmospheric conditions in the gravity field. The turbulent energy dissipation rate was selected as $\varepsilon = 50$ W/kg, turbulent Reynolds number, calculated on Taylor micro scale, was set as $Re_{\lambda} = 300$. Estimation the particles relaxation time, sedimentation velocity and other parameters of turbulence is conducted with the help of [10]. In Fig. 10(a) one can see picture of turbulent relative velocities between particles of different sizes. Relative velocity between small droplets decreases to zero. For large droplets with sufficiently inertia and velocity slip the intensity of turbulent motion decrease. For two droplets with noticeable difference in sizes the intensity of relative motion is determined by intensity of chaotic motion of droplet with lesser diameter. In Fig. 10(b) is shown the nondimensional turbulent relative velocities between droplets with equal diameters. We can see the common tendencies. Motion of small droplets are well correlated, increase in the droplet sizes leads to increasing turbulent relative velocity between droplets. For large droplets relative velocity decreases, this fact is explained as a result of general reduction chaotic motion of the droplets.

Coefficient of relative turbulent diffusion of two droplets in the atmospheric conditions is shown on the Fig. 11.



Fig. 10. Relative turbulent velocities between two particles with various diameters (a). Relative turbulent velocities between two particles with equal diameters (b).



Fig. 11. Coefficient of relative turbulent diffusion between two particles with various diameters (a). Coefficient of relative turbulent diffusion between two particles with equal diameters (b).

Fig. 11(a) presents the coefficient of relative turbulent diffusion for droplets with different diameters. For very small droplets the coefficient of diffusion tends to zero value. Turbulent motion of droplets with greater diameters is less correlated and coefficient of turbulent relative diffusion increases. The influence of crossing trajectory effect on coefficient of relative turbulent diffusion is noticeable on Fig. 11(b). For larger droplets the coefficient of diffusion decreases because sufficient velocities slip due to gravitational field.

7. Conclusions

PDF approach for describing relative motion of particles with different sizes in inhomogeneous turbulence is created. The closed system of equation for mass transfer due to turbulent relative motion of particles is obtained. Closed expressions for intensity of chaotic relative motions of particles and relative turbulent diffusion are found. Influence of particles relaxation times, averaged velocity slips and turbulence parameters is investigated. The results of calculation illustrate the main features of relative motion of particles in the wide diapason of dispersed phase parameters.

In the next paper in the atmospheric conditions coagulation process of particles with different diameters will be considered.

Appendix A. Probability density functions for particles displacement

For describing transfer of two particles in the turbulent gas flow we introduce indicator function, which represent the particles coordinates $\mathbf{x}'_{\alpha}, \mathbf{x}'_{\beta}$ and velocities $\mathbf{V}'_{\alpha}, \mathbf{V}'_{\beta}$ at moment of time t' provided that at the previous moment of time t'' < t' the coordinates and velocities of the particles was $\mathbf{x}''_{\alpha}, \mathbf{x}''_{\beta}$ and $\mathbf{V}''_{\alpha}, \mathbf{V}''_{\beta}$ respectively

$$\begin{aligned} G_{\alpha\beta}(\mathbf{x}'_{\alpha},\mathbf{V}'_{\alpha},\mathbf{x}'_{\beta},\mathbf{V}'_{\beta},t'|\mathbf{x}''_{\alpha},\mathbf{V}''_{\alpha},\mathbf{x}''_{\beta},\mathbf{V}''_{\beta},t'') \\ &= \delta\big(\mathbf{x}'_{\alpha}-\mathbf{X}^{(\mathrm{p})}_{\alpha}(t')\big)\delta\big(\mathbf{V}'_{\alpha}-\mathbf{V}^{(\mathrm{p})}_{\alpha}(t')\big)\delta\big(\mathbf{x}''_{\alpha}-\mathbf{X}^{(\mathrm{p})}_{\alpha}(t'')\big) \\ &\times \delta\big(\mathbf{V}''_{\alpha}-\mathbf{V}^{(\mathrm{p})}_{\alpha}(t'')\big)\times\delta\big(\mathbf{x}'_{\beta}-\mathbf{X}^{(\mathrm{p})}_{\beta}(t')\big) \\ &\times \delta\Big(\mathbf{V}'_{\beta}-\mathbf{V}^{(\mathrm{p})}_{\beta}(t')\Big)\delta(\mathbf{x}''_{\beta}-\mathbf{X}^{(\mathrm{p})}_{\beta}(t''))\delta\Big(\mathbf{V}''_{\beta}-\mathbf{V}^{(\mathrm{p})}_{\beta}(t'')\Big). \end{aligned}$$

After averaging function $G_{\alpha\beta}$ over the ensemble of turbulent realizations we obtain probability of particles transfer from initial conditions $\mathbf{x}''_{\alpha}, \mathbf{x}''_{\beta}, \mathbf{V}''_{\alpha}, \mathbf{V}''_{\beta}$ at the moment of time t'' to the subsequent point $\mathbf{x}'_{\alpha}, \mathbf{x}'_{\beta}, \mathbf{V}'_{\alpha}, \mathbf{V}'_{\beta}$ at the moment of time t' > t''.

Equation for indicator function $G_{\alpha\beta}$ follows from equations for particles motion in Lagrange variables (1) and have the form

$$\frac{\partial G_{\alpha\beta}}{\partial t} + V_{\alpha,k} \frac{\partial G_{\alpha\beta}}{\partial x_{\alpha,k}} + V_{\beta,k} \frac{\partial G_{\alpha\beta}}{\partial x_{\beta,k}} \\
+ \frac{\partial}{\partial V_{\alpha,k}} \left[\frac{U_k(\mathbf{x}_{\alpha}, t) + W_{\alpha,k} - V_{\alpha,k}}{\tau_{\alpha}} G_{\alpha\beta} \right] \\
+ \frac{\partial}{\partial V_{\beta,k}} \left[\frac{U_k(\mathbf{x}_{\beta}, t) + W_{\beta,k} - V_{\beta,k}}{\tau_{\chi}} G_{\alpha\beta} \right] = 0.$$
(A.1)

Initial condition for function $G_{\alpha\beta}$ have a standard form

$$G_{\alpha\beta}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, \mathbf{x}'_{\beta}, \mathbf{V}'_{\beta}, t'' | \mathbf{x}''_{\alpha}, \mathbf{V}''_{\alpha}, \mathbf{x}''_{\beta}, \mathbf{V}''_{\beta}, t'') = \delta(\mathbf{x}'_{\alpha} - \mathbf{x}''_{\alpha}) \delta(\mathbf{V}_{\alpha} - \mathbf{V}^{\circ}_{\alpha}) \delta(\mathbf{x}'_{\beta} - \mathbf{x}''_{\beta}) \delta(\mathbf{V}'_{\beta} - \mathbf{V}''_{\beta}).$$
(A.2)

Solution of Eq. (A.1) with initial condition (A.2) may be factorized as

$$G_{\alpha\beta}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, \mathbf{x}'_{\beta}, \mathbf{V}'_{\beta}, t' | \mathbf{x}''_{\alpha}, \mathbf{V}''_{\alpha}, \mathbf{x}''_{\beta}, \mathbf{V}''_{\beta}, t'')$$

= $G_{\alpha}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t' | \mathbf{x}''_{\alpha}, \mathbf{V}''_{\alpha}, t'') \times G_{\beta}(\mathbf{x}'_{\beta}, \mathbf{V}'_{\beta}, t' | \mathbf{x}''_{\beta}, \mathbf{V}''_{\beta}, t'').$
(A.3)

Here the two factors in the left hand side (A.3) are indicator functions for each particle. For example indicator function in (A.3) for α particle have the following form

$$\begin{aligned} G_{\alpha}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t' | \mathbf{x}''_{\alpha}, \mathbf{V}''_{\alpha}, t'' |) \\ &= \exp\left(3\frac{t'-t''}{\tau_{\alpha}}\right) \delta\left\{\mathbf{x}'_{\alpha} - \mathbf{x}''_{\alpha} + \tau_{\alpha}(\mathbf{V}'_{\alpha} - \mathbf{V}''_{\alpha}) - \mathbf{W}_{\alpha}(t'-t'') \right. \\ &\left. - \int_{t''}^{t'} \mathbf{U}(\mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t'), \zeta) \, \mathrm{d}\zeta\right\} \\ &\times \delta\left\{\mathbf{V}'_{\alpha} \exp\left(\frac{t'-t''}{\tau_{\alpha}}\right) - \mathbf{V}''_{\alpha} + \mathbf{W}_{\alpha}\left[1 - \exp\left(\frac{t'-t''}{\tau_{\alpha}}\right)\right] \\ &\left. - \frac{1}{\tau_{\alpha}}\int_{t''}^{t'} \exp\left(\frac{\zeta - t''}{\tau_{\alpha}}\right) \mathbf{U}(\mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t'), \zeta) \, \mathrm{d}\zeta\right\}. \end{aligned}$$
(A.4)

In (A.4) conditional displacement of particle $\mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, t')$ in integral terms means that particle travels during time interval $\zeta - t''$ under condition that at the last moment of time t' coordinate and velocity of the particle will be $\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}$

$$\begin{split} \mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t') &= \mathbf{x}'_{\alpha} + \tau_{\alpha} \mathbf{V}'_{\alpha} \bigg[1 - \exp\left(-\frac{t' - \zeta}{\tau_{\alpha}}\right) \bigg] \\ &+ \int_{\zeta}^{t'} \bigg[1 - \exp\left(-\frac{\zeta - \zeta'}{\tau_{\alpha}}\right) \bigg] \\ &\times \mathbf{U}(\mathbf{X}_{\alpha}(\zeta' | \mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t'), \zeta') \, \mathrm{d}\zeta'. \end{split}$$

In expression (A.4) we introduce the fluctuating and averaged velocities of particle and carrier phase $V_{\alpha} = \langle V_{\alpha} \rangle + v_{\alpha}$, $U_{\alpha} = \langle U_{\alpha} \rangle + u_{\alpha}$. We become attached to the coordinate frame moving with averaged velocity of carrier phase $x \rightarrow x - \langle U \rangle t$. In the coordinate frame fixed with averaged velocity of gas the indicator function (A.4) for particle velocity fluctuations and its relative displacement turn out

$$G_{\alpha}(\mathbf{x}'_{\alpha}, \mathbf{v}'_{\alpha}, t' | \mathbf{x}''_{\alpha}, \mathbf{v}''_{\alpha}, t'') = \exp\left(3\frac{t'-t''}{\tau_{\alpha}}\right) \times \delta\left\{\mathbf{x}'_{\alpha} - \mathbf{x}''_{\alpha} - \tau_{\alpha}\mathbf{v}'_{\alpha}\left[\exp\left(\frac{t'-t''}{\tau_{\alpha}}\right) - 1\right] - \mathbf{W}_{\alpha}(t'-t'') - \int_{t''}^{t'} \left[1 - \exp\left(\frac{\zeta - t''}{\tau_{\alpha}}\right)\right] \mathbf{u}(\mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, \mathbf{v}'_{\alpha}, t'), \zeta) d\zeta\right\} \times \delta\left\{\mathbf{v}'_{\alpha} \exp\left(\frac{t'-t''}{\tau_{\alpha}}\right) - \mathbf{v}''_{\alpha} - \frac{1}{\tau_{\alpha}}\int_{t''}^{t} \exp\left(\frac{\zeta - t''}{\tau_{\alpha}}\right) \times \mathbf{u}(\mathbf{X}_{\alpha}(\zeta | \mathbf{x}'_{\alpha}, \mathbf{v}'_{\alpha}, t'), \zeta) d\zeta\right\}.$$
(A.5)

After integration the expression (A.5) over the velocity space \mathbf{v}'_{α} we obtain the indicator function for displacement of α th particle on distance $\mathbf{z}_{\alpha} = \mathbf{x}'_{\alpha} - \mathbf{x}''_{\alpha}$ during interval of time s = t' - t'' under condition that at the time point t''the initial particle velocity is \mathbf{v}''_{α}

$$\begin{aligned} G_{\alpha}(\mathbf{x}_{\alpha}', t' | \mathbf{x}_{\alpha}'', \mathbf{v}_{\alpha}'', t'') &= G_{\alpha}(\mathbf{z}_{\alpha}, s | \mathbf{v}_{\alpha}'') \\ &= \delta \bigg\{ \mathbf{z}_{\alpha} - \tau_{\alpha} \mathbf{v}_{\alpha}'' \bigg[1 - \exp\left(-\frac{s}{\tau_{\alpha}}\right) \bigg] - \mathbf{W}_{\alpha} s \\ &- \int_{t''}^{t'} \bigg[1 - \exp\left(-\frac{t' - \zeta}{\tau_{\alpha}}\right) \bigg] \\ &\times \mathbf{u}(\mathbf{X}_{\alpha}(\zeta | \mathbf{x}_{\alpha}', \mathbf{v}_{\alpha}', t'), \zeta) \, \mathrm{d}\zeta \bigg\}. \end{aligned}$$
(A.6)

Probability density function of particle transfer $\langle G_{\alpha}(\mathbf{z}_{\alpha},s) \rangle$ follows from (A.6) after averaging over the ensemble of turbulent realization and ensemble of velocity fluctuations of particle. With the help of distribution function for particle velocity fluctuations $\varphi_{\alpha\beta}(\mathbf{v}'_{\alpha},\mathbf{v}'_{\beta})$ (27) we obtain

$$\langle G_{\alpha}(\mathbf{z}_{\alpha},s)\rangle = \int d\mathbf{v}_{\beta}'' \int \left\langle G_{\alpha}(\mathbf{z}_{\alpha},s|\mathbf{v}_{\alpha}'')\right\rangle_{u} \varphi_{\alpha\beta}(\mathbf{v}_{\alpha}'',\mathbf{v}_{\beta}') d\mathbf{v}_{\alpha}''. \quad (A.7)$$

The subscript u in (A.7) denotes the averaging over the ensemble of turbulent realization of gas velocity fluctuations.

With a view of profitability of the manuscript we write down formulas for probability density functions of particle transfer in one-dimensional representation.

After integration (A.6) and (A.7) (see Appendix B) we obtain the following formula for probability density function of particle turbulent transfer

$$\langle G_{\alpha}(z_{\alpha},s)\rangle = \frac{1}{\sqrt{2\pi\Delta_{\alpha}^2}} \exp\left[-\frac{(z_{\alpha}-W_{\alpha}s)^2}{2\Delta_{\alpha}^2}\right].$$
 (A.8)

Here Δ_{α}^2 is square of random displacement of particle. Random displacement of particle depends on time *s* and is a sum of particle transition due to turbulent motion of energetic eddies and inertial transfer of particle with random velocity in the previous moment of time

$$\Delta_{\alpha}^{2}(s) = \Lambda_{\alpha}^{2}(s) + \lambda_{\alpha}^{2}(s).$$
(A.9)

The first term in the right hand side in (A.9) represents length of inertial transfer of α th particle

$$\Lambda_{\alpha}^{2}(s) = \tau_{\alpha}^{2} \langle v_{\alpha}^{2} \rangle \left[1 - \exp\left(-\frac{s}{\tau_{\alpha}}\right) \right]^{2}, \tag{A.10}$$

where $\langle v_{\alpha}^2 \rangle$ is averaged square of particle velocity fluctuations.

Second term in the right hand side in (A.9) approximate averaged square length of distance on which the particle is transferred by turbulent eddies of carrier phase

$$\lambda_{\alpha}^{2}(s) = \left[1 - \exp\left(-\frac{s}{\tau_{\alpha}}\right)\right]^{2} T_{\rm E}^{2} \langle u^{2} \rangle, \qquad (A.11)$$

where $\langle u^2 \rangle$ is averaged square of gas velocity fluctuations.

For particle with small inertia $(\tau_{\alpha} \ll T_{\rm E})$ we have $\langle v_{\alpha}^2 \rangle \approx \langle u^2 \rangle$, and from ((A.9)–(A.11)) one can see, that $\Lambda_{\alpha}^2 \rightarrow 0$. During the life time of energetic eddies $T_{\rm E}$ the square of particle transfer is $\Lambda_{\alpha}^2 \approx T_{\rm E}^2 \langle u^2 \rangle$. We can conclude that inertia less particle is transferred only by energetic turbulent eddies. The turbulent parameters of inertia less particle can be used for estimation Lagrange correlation function.

For inertial particle $(\tau_{\alpha} \gg T_{\rm E})$ intensity of the particle turbulent motion is lesser than intensity of carrier phase turbulence $\langle v_{\alpha}^2 \rangle \approx \langle u^2 \rangle T_{\rm E} / \tau_{\alpha}$. For that case $\Lambda_{\alpha}^2 \approx T_{\rm E}^2 \langle u^2 \rangle$ $(T_{\rm E} / \tau_{\alpha}), \ \lambda_{\alpha}^2 \approx T_{\rm E}^2 \langle u^2 \rangle (T_{\rm E} / \tau_{\alpha})^2$ and $\Lambda_{\alpha}^2 \approx \Lambda_{\alpha}^2 \to 0$. We conclude that relative chaotic motion of inertial particles tends to zero and $\langle G_{\alpha} (z_{\alpha},s) \rangle \to \delta(z_{\alpha} - W_{\alpha}s)$. For determination the probability density function of relative transfer of two particles we consider indicator function (A.3) which is depends on initial positions of particles $\mathbf{x}^{\circ}_{\alpha}, \mathbf{x}^{\circ}_{\beta}$ and initial velocities $\mathbf{v}^{\circ}_{\alpha}, \mathbf{v}^{\circ}_{\beta}$ at the previous moment of time $t^{\circ} \ll t'$, t''

$$G_{\alpha\beta}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, \mathbf{x}'_{\beta}, \mathbf{V}'_{\beta}, t' | \mathbf{x}^{\circ}_{\alpha}, \mathbf{V}^{\circ}_{\alpha}, \mathbf{x}^{\circ}_{\beta}, \mathbf{V}^{\circ}_{\beta}, t^{\circ}) = G_{\alpha}(\mathbf{x}'_{\alpha}, \mathbf{V}'_{\alpha}, t' | \mathbf{x}^{\circ}_{\alpha}, \mathbf{V}^{\circ}_{\alpha}, t^{\circ}) \times G_{\beta}(\mathbf{x}'_{\beta}, \mathbf{V}'_{\beta}, t' | \mathbf{x}^{\circ}_{\beta}, \mathbf{V}^{\circ}_{\beta}, t^{\circ})$$
(A.12)

After integration the expression (A.12) over velocity space $\mathbf{v}'_{\alpha}, \mathbf{v}'_{\beta}$ we obtain the indicator function which presents relative distance $\mathbf{z}'_{\alpha\beta} = \mathbf{x}'_{\alpha} - \mathbf{x}'_{\beta}$ between two particles at the moment of time t' provided, that in the previous moment of time t° initial relative distance between particles were $\mathbf{y}^{\circ}_{\alpha\beta} = \mathbf{x}^{\circ}_{\alpha} - \mathbf{x}^{\circ}_{\beta}$ and initial particles velocities were $\mathbf{v}^{\circ}_{\alpha}$ and $\mathbf{v}^{\circ}_{\beta}$ $G_{\alpha\beta}(\mathbf{x}'_{\alpha}, \mathbf{x}'_{\beta}, t' | \mathbf{x}^{\circ}_{\alpha}, \mathbf{v}^{\circ}_{\alpha}, \mathbf{x}^{\circ}_{\beta}, t^{\circ})$

$$= G_{\alpha}(\mathbf{y}'_{\alpha}, t' | \mathbf{v}^{\circ}_{\alpha}, t^{\circ}) G_{\beta}(\mathbf{y}'_{\beta}, t' | \mathbf{v}^{\circ}_{\beta}, t^{\circ})$$
$$= G_{\alpha\beta}(\mathbf{z}'_{\alpha\beta}, t' | \mathbf{y}^{\circ}_{\alpha\beta}, \mathbf{v}^{\circ}_{\alpha}, \mathbf{v}^{\circ}_{\beta}, t^{\circ}), \qquad (A.13)$$

where $\mathbf{y}'_{\alpha} = \mathbf{x}'_{\alpha} - \mathbf{x}^{\circ}_{\alpha}$ and $\mathbf{y}'_{\beta} = \mathbf{x}'_{\beta} - \mathbf{x}^{\circ}_{\beta}$ are lengths of particles path from initial coordinates.

Probability density function of relative displacement of two particles $\left\langle G_{\alpha\beta}(\mathbf{z}'_{\alpha\beta}, t'|\mathbf{y}^{\circ}_{\alpha\beta}, t^{\circ}) \right\rangle$ follows from expression (A.13) after averaging over the ensemble of turbulent realization of gas velocities and velocities distribution of two particles $\varphi_{\alpha\beta}(\mathbf{v}^{\circ}_{\alpha}, \mathbf{v}^{\circ}_{\beta})$

$$\left\langle G_{\alpha\beta}(\mathbf{z}_{\alpha\beta}', t' | \mathbf{y}_{\alpha\beta}^{\circ}, t^{\circ}) \right\rangle = \int \mathbf{d}\mathbf{v}_{\beta}^{\circ} \int \left\langle G_{\alpha\beta}(\mathbf{z}_{\alpha\beta}', t' | \mathbf{y}_{\alpha\beta}^{\circ}, \mathbf{v}_{\alpha}^{\circ}, \mathbf{v}_{\beta}^{\circ}, t^{\circ}) \right\rangle_{u} \\ \times \varphi_{\alpha\beta}(\mathbf{v}_{\alpha}^{\circ}, \mathbf{v}_{\beta}^{\circ}) \mathbf{d}\mathbf{v}_{\beta}^{\circ}.$$
(A.14)

After calculation (A.14) (see Appendix B) we obtain the following expression

$$\left\langle G_{\alpha\beta}(z_{\alpha\beta},t'|y_{\alpha\beta}^{\circ},t^{\circ})\right\rangle = \frac{1}{\sqrt{2\pi\widetilde{\Delta}_{\alpha\beta}^{2}}} \exp\left\{-\frac{\left[z_{\alpha\beta}-y_{\alpha\beta}^{\circ}-W_{\alpha\beta}(t'-t^{\circ})\right]^{2}}{2\widetilde{\Delta}_{\alpha\beta}^{2}}\right\}.$$
 (A.15)

Here $W_{\alpha\beta} = |W_{\alpha} - W_{\beta}|$ is module of relative averaged velocities of particles due to mass forces, and $\tilde{\Delta}_{\alpha\beta}^2$ is averaged square of total length of distance between particles

$$\widetilde{\varDelta}^2_{\alpha\beta} = \varDelta^2_{\alpha\beta} + \lambda^2_{\alpha\beta}.$$

The expression for square of relative distance between particles due to their inertia have the form

$$A_{\alpha\beta}^{2} = \tau_{\alpha}^{2} \langle v_{\alpha}^{2} \rangle + \tau_{\beta}^{2} \langle v_{\beta}^{2} \rangle - 2\rho_{\alpha\beta}\tau_{\alpha}\tau_{\beta}\sqrt{\langle v_{\alpha}^{2} \rangle \langle v_{\beta}^{2} \rangle}.$$
(A.16)

Expression (A.16) is obtained for interval of time $t' - t^{\circ} \gg \tau_{\alpha}, \tau_{\beta}$. The approximation of square of relative distance between particles due to motion with turbulent energy containing eddies have the following form

$$\lambda_{\alpha\beta}^{2} = T_{\rm E}^{2} \left(\mathrm{e}^{-\frac{T_{\rm E}}{\tau_{\alpha}}} - \mathrm{e}^{-\frac{T_{\rm E}}{\tau_{\beta}}} \right)^{2} \langle u^{2} \rangle. \tag{A.17}$$

For inertia less particles τ_{α} , $\tau_{\beta} \ll T_{\rm E}$ the dispersion of relative distance between particles aspires to zero $\tilde{\Delta}_{\alpha\beta}^2 \rightarrow 0$. This result coincides with conclusion that correlation coefficient $\rho_{\alpha\beta}$ (28) for inertia less particles tends to unity, and there no relative velocity between particles. For inertial particles $\tau_{\alpha} \approx \tau_{\beta} \gg T_{\rm E}$ we can see from (A.17), that the parameter $\lambda_{\alpha\beta}^2 \approx T_{\rm E}^2 \langle u^2 \rangle (T_{\rm E}/\tau_{\alpha})^3$. Inertial relative transfer between particles with sufficient inertia (A.16) becomes larger than integral scale of Euler gas velocity correlation $\Lambda_{\alpha\beta}^2 \approx T_{\rm E}^2 \langle u^2 \rangle (\tau_{\alpha}/T_{\rm E}) \gg L_{\rm E}^2$. This fact reflects the statement, that inertial particles save their dynamic information on distances, which is larger than in the case of small inertia particles.

For $z'_{\alpha\beta} = y_{\alpha\beta} = x_{\alpha} - x_{\beta}$ the expression (A.15) can be considered as probability density distribution of initial distance between particles $y^{\circ}_{\alpha\beta} = x^{\circ}_{\alpha} - x^{\circ}_{\beta}$ provided, that at the moment of time *t* the relative distance between particles will be $y_{\alpha\beta}$.

For current time ξ , which satisfies the conditions $t^{\circ} < \xi \leq t$ and $\xi - t^{\circ} \gg \tau_{\alpha}, \tau_{\beta}$, expression for probability density function of relative distance between particles is analogous (A.15)

$$\left\langle G_{\alpha\beta} \left(Y_{\alpha\beta}, \xi | y_{\alpha\beta}^{\circ}, t^{\circ} \right) \right\rangle = \frac{1}{\sqrt{2\pi \widetilde{\Delta}_{\alpha\beta}^{2}}} \exp\left\{ -\frac{\left[Y_{\alpha\beta} - y_{\alpha\beta}^{\circ} - \boldsymbol{W}_{\alpha\beta}(\xi - t^{\circ}) \right]^{2}}{2 \widetilde{\Delta}_{\alpha\beta}^{2}} \right\}, \quad (A.18)$$

where $Y_{\alpha\beta}$ is relative distance between particles at the moment of time ξ .

Expression for probability density function of relative distance between particles is $\mathbf{Y}_{\alpha\beta}$ at the time ξ provided, that relative distance between particles at the moment of time *t* will be $y_{\alpha\beta}$ follows from (A.15) and (A.18) $\langle G(\mathbf{x}_{\alpha}, t | \mathbf{Y}_{\alpha\beta}, \xi) \rangle$

$$G(\mathbf{y}_{\alpha\beta}, t | \mathbf{Y}_{\alpha\beta}, \zeta) \rangle = \int \left\langle G_{\alpha\beta}(\mathbf{y}_{\alpha\beta}, t | \mathbf{y}_{\alpha\beta}^{\circ}, t^{\circ}) \right\rangle \left\langle G_{\alpha\beta}(\mathbf{Y}_{\alpha\beta}, \zeta | \mathbf{y}_{\alpha\beta}^{\circ}, t^{\circ}) \right\rangle d\mathbf{y}_{\alpha\beta}^{\circ}.$$
(A.19)

After calculation (A.19) in one dimension (see Appendix B) we obtain expression for probability density function of relative distance between particles $Y_{\alpha\beta}$ at the moment of time ξ under condition that distance between particles at the moment of time t is equal $y_{\alpha\beta}$

$$\left\langle G_{\alpha\beta}\left(y_{\alpha\beta},t|Y_{\alpha\beta},\xi\right)\right\rangle = \frac{1}{\sqrt{2\pi\Delta_{\alpha\beta}^2}} \exp\left\{-\frac{\left[Y_{\alpha\beta}-y_{\alpha\beta}-\boldsymbol{W}_{\alpha\beta}(t-\xi)\right]^2}{2\Delta_{\alpha\beta}^2}\right\},\tag{A.20}$$

$$\Delta_{\alpha\beta}^2 = 2\widetilde{\Delta}_{\alpha\beta}^2 = 2(\Lambda_{\alpha\beta}^2 + \lambda_{\alpha\beta}^2). \tag{A.21}$$

From (A.20) one can see, that increasing relative distance between particles $y_{\alpha\beta}$ and module of particles relative velocities $W_{\alpha\beta}$ decrease function $\langle G(\mathbf{y}_{\alpha\beta}t|\mathbf{Y}_{\alpha\beta},\xi)\rangle$ and diminish conditional gas velocity correlation $\Psi_{\beta|\alpha}^{(p)}(s, y_{\alpha\beta})$ in (45). It leads to reduction conditional response function $f_{\beta|\alpha}$, that in terns decrease correlation coefficient between particles $\rho_{\alpha\beta}$ (28). It is necessary to note the self-coordinated character of the expressions (A.20) and (A.21). If we consider one particle relative distance is absent $y_{\alpha\beta} = 0$, $\tau_{\alpha} = \tau_{\beta}$ and correlation coefficient in (A.16) is $\rho_{\alpha\beta} = 1$. The square of dispersion $\Delta^2_{\alpha\beta} = 0$ (see expression (A.21)) and function (A.20) $\langle G_{\alpha\beta}(y_{\alpha\beta},t|Y_{\alpha\beta},\xi) \rangle \rightarrow \delta(Y_{\alpha\beta})$. From expressions (46) and (47) one can see, that response functions $f_{\beta|\alpha} = f_{\alpha|\beta} \rightarrow f_{\alpha}$.

Appendix B. Calculation of some integrals

We present the results of calculation of some integrals used in the paper. At determination the probability density function of particles relative distance it is used the following expression

$$G(Y) = \int_{-\infty}^{\infty} \mathrm{d}v_{\alpha} \int_{-\infty}^{\infty} \varphi_{\alpha\beta}(v_{\alpha}, v_{\beta}) \delta(Y - (av_{\alpha} - bv_{\beta})) \,\mathrm{d}v_{\beta},$$
(B.1)

where function $\varphi_{\alpha\beta}(v_{\alpha},v_{\beta})$ present the probability density function of particles velocity distribution in one dimension

$$\begin{split} \varphi_{\alpha\beta}(v_{\alpha}, v_{\beta}) &= \frac{1}{2\pi\sqrt{(1 - \rho_{\alpha\beta}^2)\sigma_{\alpha}\sigma_{\beta}}} \\ &\times \exp\left[-\frac{1}{2(1 - \rho_{\alpha\beta}^2)}\left(\frac{v_{\alpha}^2}{\sigma_{\alpha}} + \frac{v_{\beta}^2}{\sigma_{\beta}} - \frac{2\rho_{\alpha\beta}v_{\alpha}v_{\beta}}{\sqrt{\sigma_{\alpha}\sigma_{\beta}}}\right)\right]. \end{split}$$
(B.2)

Result of substitution (B.2) into (B.1) has the form of Gaussian distribution

$$G(Y) = \frac{1}{\sqrt{2\pi \Delta_{\alpha\beta}^2}} \exp\left(-\frac{Y^2}{2\Delta_{\alpha\beta}^2}\right),\tag{B.3}$$

where $\varDelta_{\alpha\beta}$ is dispersion of random distance between particles

$$\Delta_{\alpha\beta}^2 = a^2 \sigma_{\alpha} + b^2 \sigma_{\beta} - 2\rho_{\alpha\beta} a b \sqrt{\sigma_{\alpha} \sigma_{\beta}}.$$
 (B.4)

We see from (B.4), that the dispersion of random distance between particles is reduced with grows the coefficient correlation $\rho_{\alpha\beta}$. For uncorrelated particles $\rho_{\alpha\beta} = 0$ square of relative particle distance $\Delta^2_{\alpha\beta}$ reach a maximum value, and distribution G(Y) (B.3) becomes wider.

The following integral appears during the calculation of probability density function of turbulent relative transfer between two particles

$$G(Y-Z) = \int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi\lambda_1^2}} \exp\left[-\frac{(Y-y)^2}{2\lambda_1^2}\right] \frac{1}{\sqrt{2\pi\lambda_2^2}}$$
$$\times \exp\left[-\frac{(Z-y)^2}{2\lambda_2^2}\right] dy$$
$$= \frac{1}{\sqrt{2\pi\Delta^2}} \exp\left[-\frac{(Y-Z)^2}{2\Lambda^2}\right], \quad \Lambda^2 = \lambda_1^2 + \lambda_2^2$$

Integral which arise at the calculation of correlation of gas phase velocity fluctuations along a particle trajectory have the form

$$\psi(L, \Delta, a) = \int_0^\infty \frac{1}{\sqrt{2\pi}\Delta} \exp\left[-\frac{(y-a)^2}{2\Delta^2}\right] \exp\left(-\frac{y}{L}\right) \mathrm{d}y.$$

After calculation we obtain the factor in the expression (49) for gas velocity correlation along the particle trajectory

$$\psi(L, \Delta, a) = \frac{1}{2} \left[\exp\left(\frac{\Delta^2}{2L^2} - \frac{a}{L}\right) \operatorname{erfc}\left(\sqrt{2}\left(\frac{\Delta}{L} - \frac{a}{\Delta}\right)\right) + \exp\left(\frac{\Delta^2}{2L^2} + \frac{a}{L}\right) \operatorname{erfc}\left(\sqrt{2}\left(\frac{\Delta}{L} + \frac{a}{\Delta}\right)\right) \right]$$
(B.5)

where $\operatorname{erfc}(x) = 1 - \operatorname{erf}(x)$, $\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt$ is standard error function.

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