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Abstract

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GEOPHYSICS

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EQUATIONS OF BROWNIAN MOTION OF AEROSOL PARTICLES

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In the course of certain atmospheric processes, as is known, Brownian diffusion of aerosol particles plays an essential role; moreover, the inertia of the particles often cannot be neglected, and therefore the usual Smoluchowski equation becomes unsuitable for studying diffusion. In ⁽¹⁾ approximate equations were obtained that take into account the inertia of aerosol particles, and in ^(2, 3) some results of calculations carried out on their basis were presented; this made it possible to reveal interesting, qualitatively new effects in the theory of diffusion: the effect of concentration growth and the effect of displacement from the trajectory of the particles of the medium, which in some cases substantially change the magnitudes of the diffusion fluxes as compared with the inertia-free approximation. Establishing the limits of applicability of the approximate equations ⁽¹⁾, as well as deriving more accurate equations, is the purpose of the present note.

We shall proceed from the Fokker-Planck equation ⁽⁴⁾

$$\frac{\partial f}{\partial t} + \left(v_i \frac{\partial}{\partial x_i} + \frac{\partial}{\partial v_i} F_i^* \right) f = \frac{1}{k^2 \text{Pe}} \frac{\partial^2}{\partial v_i^2} f, \quad (1)$$

$$= f(\mathbf{r}, \mathbf{v}, t), \quad \mathbf{F}^* = \frac{1}{k} (-\mathbf{v} + \mathbf{u} + \mathbf{F}), \quad k = mu_\infty/b, \quad \text{Pe} = mu_\infty^2/k_B T.$$

Here f is the distribution function of particles in phase space; $\mathbf{u}$ is the dimensionless velocity of the medium; $\mathbf{F}$ is the dimensionless external force acting on a particle; m is the particle mass; b is the friction coefficient; u_∞ and l are the characteristic velocity and size of the flow; k_B is Boltzmann's constant; T is the temperature of the medium. In what follows it is assumed that the vector field $\mathbf{u} + \mathbf{F}$ is solenoidal.

To close the chain of equations for the moments of the function f , obtained from equation (1), we shall use the following hypothesis. Investigation of the

motion of free particles in a medium for which $\mathbf{u} = \text{const}$ shows that the particles acquire a velocity equal to the velocity of the medium over a time $\tau^* \sim k$ (τ^* is the relaxation time). Therefore it may be assumed that, for $t \gg \tau^*$, in a system of aerosol particles in certain regions Q^* of configuration space, local equilibrium is established, and the further process proceeds with preservation of local equilibrium. Apparently, Q^* will include the entire configuration volume occupied by the flow, except for thin layers in front of discontinuities, including the surfaces of obstacle bodies. Let us further assume that, whatever the initial distribution of particles in the system may have been, for $t \gg \tau^*$ the distributions over velocities and coordinates are statistically independent and the distribution over velocities has an approximately locally Maxwellian form:

$$f \sim A n e^{-\frac{1}{2} k \text{Pe} |\mathbf{v} - \langle \mathbf{v} \rangle|^2}, \quad A = \left(\frac{k \text{Pe}}{2\pi} \right)^{3/2}, \quad t \gg \tau^*, \quad \mathbf{r} \in Q^*, \quad (2)$$

where n is the concentration of aerosol particles, $\langle \mathbf{v} \rangle$ is their mean velocity. It is obvious that the approximate relation (2) will be satisfied when

n and $\langle \mathbf{v} \rangle$ either do not depend on t at all (the stationary case), or this dependence is so weak that the time of their appreciable change τ^{**} substantially exceeds τ^* (the quasi-stationary case).

It is natural to call the system of aerosol particles described by a distribution function of type (2) an aerosol fluid. It is immediately clear that this function determines only the transfer of the characteristics of the fluid associated with concentration gradients; transfer caused by gradients of the velocity field, i.e., the “viscosity” of the Brownian fluid, is completely neglected. If (2) is substituted into (1), then one can establish that the exact particular solution of the Fokker–Planck equation would be the function (2) under the condition that its right-hand side has the form

$$\frac{1}{k^2 \text{Pe}} \left[\delta_{ij} + \frac{1}{2} k \left(\frac{\partial \langle v_i \rangle}{\partial x_j} + \frac{\partial \langle v_j \rangle}{\partial x_i} \right) \right] \frac{\partial^2 f}{\partial v_i \partial v_j}.$$

Since the right-hand side in (2) is responsible for diffusion and, consequently, has order $O(\varphi(\text{Pe}))$, where $\varphi(\text{Pe}) \rightarrow 0$ as $\text{Pe} \rightarrow \infty$, we arrive at the conclusion that (2) is the principal part of the asymptotic expansion of some particular solution of (1) in the parameter $\lambda = \varphi(\text{Pe}) k$ for $k \ll 1$ and $\text{Pe} \gg 1$.

Integrating (1) over the “velocity” volume, and then multiplying by $\mathbf{v}$ and again integrating over the “velocity” volume and using (2), we obtain the following equations of motion of the aerosol fluid:

$$\frac{\partial n}{\partial t} + \text{div } n \langle \mathbf{v} \rangle = 0, \quad (3)$$

$$k \frac{\partial \langle \mathbf{v} \rangle}{\partial t} + k(\langle \mathbf{v} \rangle \nabla) \langle \mathbf{v} \rangle + \langle \mathbf{v} \rangle = \mathbf{u} + \mathbf{F} - \frac{1}{\text{Pe}} \nabla \ln n + O(\lambda).$$

Since, in the main, in the study of processes in the atmosphere for Brownian aerosol particles the conditions $k \ll 1$ and $\text{Pe} \gg 1$ are satisfied, the equations obtained for $t \gg \tau^*$, $\tau^{**} \gg \tau^*$, $\mathbf{r} \in Q^*$ are sufficiently accurate. In the particular case when $\mathbf{u} + \mathbf{F} = 0$, equations (3) coincide with the equations given in (5).

Represent $\langle \mathbf{v} \rangle$ in the form $\langle \mathbf{v} \rangle = \mathbf{v}_1 + \mathbf{v}_2$. Let $\mathbf{v}_1$ satisfy the equation

$$k \partial \mathbf{v}_1 / \partial t + k(\mathbf{v}_1 \nabla) \mathbf{v}_1 + \mathbf{v}_1 = \mathbf{u} + \mathbf{F}, \quad (4)$$

i.e., the equation of purely inertial motion of particles in the force field $\mathbf{u} + \mathbf{F}$. Then $\mathbf{v}_2$ will satisfy the equation

$$k \frac{\partial \mathbf{v}_2}{\partial t} + k(\mathbf{v}_2 \nabla) \mathbf{v}_2 + \mathbf{v}_2 = -\frac{1}{\text{Pe}} \nabla \ln n - k[(\mathbf{v}_1 \nabla) \mathbf{v}_2 + (\mathbf{v}_2 \nabla) \mathbf{v}_1]. \quad (5)$$

Consequently, in the diffusion of aerosol particles there appears an addition to the purely inertial velocity $\mathbf{v}_1$, caused by the influence of the generalized diffusion force

$$\mathbf{F}_d = -\frac{1}{\text{Pe}} \nabla \ln n$$

and by interference of inertial and diffusion motion, which is equivalent to the appearance in the system of an effective force

$$\mathbf{F}_e = -k[(\mathbf{v}_1 \nabla) \mathbf{v}_2 + (\mathbf{v}_2 \nabla) \mathbf{v}_1].$$

We note that $\mathbf{F}_d$ is proportional only to the concentration gradient because of the complete neglect, adopted above, of the viscosity of the aerosol fluid. By virtue of the assumption of solenoidality of the field $\mathbf{u} + \mathbf{F}$, under the corresponding boundary conditions we obtain that $\nabla \ln n = O(k)$ as $k \rightarrow 0$ and $\text{Pe} \rightarrow \infty$; consequently, $\mathbf{F}_d = O(k/\text{Pe})$. Taking into account the viscosity of the aerosol fluid will lead to the appearance in $\mathbf{F}_d$ of a term that will be proportional to $\psi(k/\text{Pe})$, where $\psi(\lambda)/\lambda \rightarrow 0$ as $\lambda \rightarrow 0$. Thus,

$$\mathbf{F}_d = -\frac{1}{\text{Pe}} \nabla \ln n + o(k/\text{Pe}), \quad k \rightarrow 0, \quad \text{Pe} \rightarrow \infty.$$

From (3) it is not difficult to obtain the following asymptotic relation:

$$\frac{\partial n}{\partial t} + \operatorname{div} n \left\{ \mathbf{u} + \mathbf{F} + k \left[\frac{\partial}{\partial t} + (\mathbf{u} + \mathbf{F} \cdot \nabla) \right] (\mathbf{u} + \mathbf{F}) + O(k^2) \right\} = \frac{1}{\operatorname{Pe}} \Delta n, \quad k \rightarrow 0. \quad (6)$$

It follows from (6) that, in the case when the second term in the braces is small compared with the first, equations (3) are equivalent to the Smoluchowski equation for the diffusion of Brownian particles in a force field. Thus, the criterion for the applicability of the inertia-free approximation is the condition

$$k |[\partial/\partial t + (\mathbf{u} + \mathbf{F} \cdot \nabla)] (\mathbf{u} + \mathbf{F})| \ll |\mathbf{u} + \mathbf{F}|. \quad (7)$$

Introducing a new velocity $\mathbf{v}^*$

$$\langle \mathbf{v} \rangle = \mathbf{v}^* - \frac{1}{\operatorname{Pe}} \nabla \ln n$$

equations (3) can be transformed into the form

$$\frac{\partial n}{\partial t} + \operatorname{div} n \mathbf{v}^* = \frac{1}{\operatorname{Pe}} \Delta n; \quad (8)$$

$$k \frac{\partial}{\partial t} \left(\mathbf{v}^* - \frac{1}{\operatorname{Pe}} \nabla \ln n \right) + k \left(\mathbf{v}^* - \frac{1}{\operatorname{Pe}} \nabla \ln n \cdot \nabla \right) \left(\mathbf{v}^* - \frac{1}{\operatorname{Pe}} \nabla \ln n \right) + \mathbf{v}^* = \mathbf{u} + \mathbf{F}. \quad (9)$$

Since (8) formally coincides with the equation of convective diffusion, it is natural to regard $\mathbf{v}^*$ as the convective velocity of the aerosol fluid, and $n \mathbf{v}^*$ as the convective flux of Brownian particles. From (9) it is seen that, under the condition

$$\frac{1}{\operatorname{Pe}} |\nabla n| \ll |n \mathbf{v}^*| \quad (10)$$

the convective velocity $\mathbf{v}^*$ becomes equal to the velocity of purely inertial motion:

$$\mathbf{v}^* \approx \mathbf{v}_1. \quad (11)$$

Equations (8) and (11) constitute the quasistationary approximation in the theory of diffusion of aerosol particles. They were obtained in ⁽¹⁾. The criterion for applicability of this approximation is condition (10). Let us note, however, that the quasistationary approximation will give satisfactory results also in the case when condition (10) is not satisfied in some small part of the flow region, but in this part the influence of particle inertia on $\mathbf{v}^*$ may be neglected. Only

for this reason can the results presented in ^(2, 3) be close to the truth, since for small k condition (10) is certainly not satisfied near the surfaces of obstacle bodies.

In ⁽¹⁾, for a potential field $\mathbf{u} + \mathbf{F}$, the quasi-inertial approximation was somewhat refined by approximately taking into account the interference of inertial and diffusive motions. We shall carry out a refinement of the same type for a nonpotential field $\mathbf{u} + \mathbf{F}$. When condition (10) is satisfied, in the first approximation we have

$$\mathbf{v}_2 \approx -\frac{1}{\text{Pe}} \nabla \ln n.$$

A further approximation can be found from (5), completely neglecting the gradients (and the time derivative) of $\mathbf{v}_2$ in comparison with the gradients of $\mathbf{v}_1$. We have

$$\mathbf{v}_2 \approx -\frac{1}{\text{Pe}} \nabla \ln n - k(\mathbf{v}_2 \nabla) \mathbf{v}_1.$$

Consequently,

$$\mathbf{v}_2 \approx -\frac{1}{\text{Pe}} \nabla \ln n \cdot \beta^{-1}, \quad \langle \mathbf{v} \rangle \approx \mathbf{v}_1 - \frac{1}{\text{Pe}} \nabla \ln n \cdot \beta^{-1},$$

where β is a tensor whose components are equal to $\delta_{ij} + k \partial v_{1j} / \partial x_i$. Substituting the value found for $\langle \mathbf{v} \rangle$ into the first equation of system (3), we obtain an equation for the concentration

$$\frac{\partial n}{\partial t} + \text{div } n \mathbf{v}_1 = \frac{1}{\text{Pe}} \text{div}(\nabla n \cdot \beta^{-1}), \quad (12)$$

which, together with equation (11), forms the system of equations of Brownian motion of aerosol particles in the quasi-inertial approximation, with approximate allowance for the interference of diffusive and inertial motions. If the field of the vector $\mathbf{u} + \mathbf{F}$ is potential, then it can be shown that the vector $\mathbf{v}_1$ will also be potential; consequently, the tensor β is symmetric, and therefore the equality

$$\nabla n \cdot \beta^{-1} = \beta^{-1} \cdot \nabla n$$

will hold, and equation (12) is transformed into the equation obtained in (1).

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