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A mechanism for the formation of aerosol concentrations in the atmosphere of Titan

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Abstract. A mechanism of formation of concentrations of light aerosol particles in the turbulent atmosphere of Titan is suggested. Formation of aerosol concentrations in the region of the temperature inversion is caused by the recently discovered effects of turbulent barodiffusion and turbulent thermal diffusion. Using the parameters of the atmosphere of Titan, the relative variation of the optical depth of the atmosphere caused by the accumulation of the aerosol particles is estimated. Assuming a steady-state distribution and averaging over the effect of coagulation, it is found, in the linear approximation, that optical depth enhancements of tens of percent may be possible. © 1997 Published by Elsevier Science Ltd

1. Introduction

The aerosols in Titan's atmosphere have a major effect on the heat balance and dynamics of that atmosphere. As a result, they have been the subject of a number of investigations. These have included laboratory simulations of the aerosols themselves (Bar-Nun *et al.*, 1988; Scattergood *et al.*, 1992), as well as theoretical computations of their light-scattering properties (Podolak and Danielson, 1977; Rannou *et al.*, 1995), their contribution to the atmospheric heat budget (McKay *et al.*, 1989) and the microphysics of their growth (Podolak and Podolak, 1980; Toon *et al.*, 1992). Recently, calculations have even begun to include crude modeling of the dynamic behavior of the aerosols (e.g. Hutzell *et al.*, 1996). In this study we present a new dynamical mechanism, based on the recently discovered effects of turbulent barodiffusion and turbulent thermal diffusion (see Elperin *et al.*, 1995, 1996a, 1997), which can have an important effect on the vertical distribution of the aerosols. As a result, it will affect the computed albedo of the satellite and, hence the quality of a given model's fit to the observations. Below, we present the theory underlying this effect and compute its magnitude for conditions relevant to Titan.

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2. The governing equations

If $n_p(t, \mathbf{r})$ is the number density of light particles in some fluid (such as the Titan atmosphere) at time t and position $\mathbf{r}$, then the evolution of this distribution is determined by the equation of convective diffusion:

$$\frac{\partial n_p}{\partial t} + \vec{\nabla} \cdot (n_p \mathbf{v}) = -\vec{\nabla} \cdot \mathbf{J} \tag{1}$$

where v is the velocity of the medium and the diffusive flux $\mathbf{J}$ in the case of heavy passive scalar particles (i.e. where the mass of a particle is much greater than the mean molecular weight of the surrounding fluid) is given by

$$\mathbf{J} = -D\left(\vec{\nabla} n_p - \frac{m_p \mathbf{g}}{\kappa T} n_p\right) \tag{2}$$

(e.g. Akhiezer and Petleminskii, 1981). Here D is the coefficient of molecular diffusion, κ is Boltzmann's constant, $\mathbf{g}$ is the acceleration of gravity, T is the temperature of the surrounding fluid, m_p is the mass of a particle and m_μ is the mass of a molecule of the surrounding fluid. The second term in the flux $\mathbf{J}$ describes the sedimentation of the passive scalar particles in a gravity field with a velocity $\mathbf{v}_s = D(m_p \mathbf{g}/\kappa T)$. This means that the velocity of these particles is given by $\mathbf{v}_p = \mathbf{v} + \mathbf{v}_s$. We consider a Brownian approximation for diffusivity of particles. In this case the coefficient of diffusion is given by

$$D = \frac{\kappa T}{6\pi a_* \rho \nu}$$

(e.g. Landau and Lifshitz, 1987), where a_* is the radius of a Brownian particle and ν is the kinematic viscosity of the carrying fluid. Note that, although equation (1) does not formally include coagulation, it is applicable to a volume averaged density of particles. This can be seen by multiplying the coagulation equation by the volume of the particles and integrating over the size distribution of the aerosols. The integral term weighted with the particle mass vanishes and we get the continuity equation (1) for the volume averaged density of particles. Thus, the equation can be seen as describing an average effect which does not take explicit account of the size distribution of the particles.

The velocity $\mathbf{v}$ and the density ρ of the medium satisfy the continuity equation

$$\frac{\partial \rho}{\partial t} + \vec{\nabla}\cdot(\rho \mathbf{v}) = 0. \tag{3}$$

In order to derive an equation for the mean mass concentration we average equation (1) over an ensemble of random velocity fluctuations. For this purpose we use the stochastic calculus which was applied in magnetohydrodynamics (Zeldovich *et al.*, 1988, 1990; Kleeorin and Rogachevskii, 1994) and for passive scalar transport in incompressible (Zeldovich *et al.*, 1988, 1990; Avellaneda and Majda, 1994) and compressible (Elperin *et al.*, 1995, 1996a,b, 1997) turbulent flows. The use of the technique described in Elperin *et al.* (1995, 1997) allows one to derive an equation for the mean field $N = \langle n_p \rangle$:

$$\frac{\partial N}{\partial t} + \vec{\nabla}\cdot((\mathbf{V}_{\rm eff} + \mathbf{v}_s)N - \hat{D}\vec{\nabla}N) = 0 \tag{4}$$

where

$$\hat{D} \equiv D_{\rm pm} = D\delta_{\rm pm} + \langle \tau u_p u_m \rangle \tag{5}$$

$$\mathbf{V}_{\rm eff} = \mathbf{V} - \langle \tau \mathbf{u}(\vec{\nabla}\cdot\mathbf{u})\rangle \tag{6}$$

and $\mathbf{v} = \mathbf{V} + \mathbf{u}$, $\mathbf{V} = \langle \mathbf{v} \rangle$ is the mean velocity and $\mathbf{u}$ is the turbulent component of the velocity, τ is the characteristic time of turbulent motions which depends on the scale of motions. Note that equation (4) is written in the form of a conservation law for the total number of particles.

The continuity equation (3) can be rewritten in the form

$$\left(1 + \frac{\tilde{\rho}}{\rho}\right)(\vec{\nabla}\cdot\mathbf{u}) = -\frac{1}{\rho}(\mathbf{u}\cdot\vec{\nabla})\rho - \frac{1}{\rho}\frac{\mathrm{d}\tilde{\rho}}{\mathrm{d}t} \tag{7}$$

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

Consider the case of low Mach numbers: $M \ll (l_0/L)^{1/2}$, where l_0 is the maximum scale of turbulent motions, L is some characteristic large scale (e.g. the inhomogeneity scale of the mean temperature or the mean density), $M = (\langle \mathbf{u}^2 \rangle)^{1/2}/c_s$ is the Mach number and c_s is the sound speed. Since the fluctuations of pressure are of the order of $\tilde{P} = c_s^2\tilde{\rho} \sim \rho\langle \mathbf{u}^2 \rangle$, the ratio $\tilde{\rho}/\rho \sim M^2 \ll 1$. Therefore, we can neglect terms $\sim \tilde{\rho}$ and equation (7) is reduced to

$$(\vec{\nabla}\cdot\mathbf{u}) \simeq -\frac{1}{\rho}(\mathbf{u}\cdot\vec{\nabla})\rho. \tag{8}$$

Using the ideal gas equation of state for the surrounding fluid for the mean fields $P = \rho\kappa T/m_\mu$ yields

$$\frac{\vec{\nabla}T}{T} - \frac{\vec{\nabla}P}{P} = -\frac{\vec{\nabla}\rho}{\rho} \equiv \vec{\lambda}. \tag{9}$$

Combining equations (4–6), (8) and (9) allows us to rewrite equation (4) in the following form

$$\frac{\partial N}{\partial t} + \vec{\nabla}\cdot(\mathbf{J}_{\rm M} + \mathbf{J}_{\rm T}) = 0 \tag{10}$$

where the turbulent flux of the particles is given by

$$\mathbf{J}_{\rm T} = -D_{\rm T}\left[\vec{\nabla}N + N\frac{\vec{\nabla}T}{T} - N\frac{\vec{\nabla}P}{P}\right] \tag{11}$$

$D_{\rm T} = u_0 l_0/3$ is the coefficient of turbulent diffusion, u_0 is the characteristic velocity in the scale l_0. The molecular flux of the particles

$$\mathbf{J}_{\rm M} = -D\left[\vec{\nabla}N + k_{\rm t}\frac{\vec{\nabla}T}{T} + k_{\rm p}\frac{\vec{\nabla}P}{P}\right] \tag{12}$$

comprises three terms: molecular diffusion ($\sim\vec{\nabla}N$), molecular thermal diffusion ($\sim k_{\rm t}\vec{\nabla}T$, where $k_{\rm t}$ is the molecular thermal diffusion ratio) and molecular barodiffusion ($\sim k_{\rm p}\vec{\nabla}P$, where $k_{\rm p}$ is the molecular barodiffusion ratio, see for example Landau and Lifshitz (1987)). Comparing the molecular (12) and turbulent (11) fluxes of particles we can interpret the new additional turbulent fluxes as fluxes caused by the effects of turbulent thermal diffusion ($\sim k_{\rm T}\vec{\nabla}T$, where $k_{\rm T} = N$ is the turbulent thermal diffusion ratio) and turbulent barodiffusion ($\sim k_{\rm P}\vec{\nabla}P$, where $k_{\rm P} = -N$ is the turbulent barodiffusion ratio).

Remarkably, the additional turbulent flux caused by the effect of turbulent thermal diffusion appears also for inertial particles advected by a turbulent flow (Elperin *et al.*, 1996a). The turbulent flux of small inertial particles of mass $m_{\rm p}$ is given by

$$\mathbf{J}_{\rm T}^{(\rm p)} = -D_{\rm T}\left[\frac{k_{\rm T}^{(\rm p)}}{T}\vec{\nabla}T + \vec{\nabla}N\right] \tag{13}$$

$$k_{\rm T}^{(\rm p)} = N\frac{3}{Pe}\left(\frac{m_{\rm p}}{m_\mu}\right)\left(\frac{T}{T_*}\right)\ln Re_* \tag{14}$$

(see Elperin *et al.*, 1996a), where $k_{\rm T}^{(\rm p)}$ is the turbulent thermal diffusion ratio of small inertial particles and $D_{\rm T}k_{\rm T}^{(\rm p)}$ is the coefficient of turbulent thermal diffusion, $Pe = u_0 l_0/D$ is the Peclet number, $Re_* = \min\{Re, Pe_{\rm T}\}$, $Re = l_0 u_0/\nu$ is the Reynolds number, $Pe_{\rm T} = l_0 u_0/\chi$ is the thermal Peclet number and χ is the coefficient of molecular thermal conductivity. Turbulent thermal diffusion of small inertial particles is caused by the correlation between temperature and velocity fluctuations of the surrounding fluid and leads to the relatively strong mean flux of small inertial particles in the direction of the regions with the minimum (or maximum) of the mean temperature of the surrounding fluid, depending on the ratio of material particle density to that of the surrounding fluid. Note that the velocity field of particles is divergent due to the finite inertia of particles advected by turbulent flow. For heavy particles (with sizes $\geqslant 1\,\mu$m) the turbulent thermal diffusion ratio $k_T^{(p)} \gg N$ at large Reynolds and Peclet numbers.

For the light particles the effect of inertia is negligible. However, compressibility of the surrounding fluid results in the new additional turbulent flux of particles caused by turbulent thermal diffusion. The additional turbulent nondiffusive fluxes of light particles can be also estimated as follows. We average equation (1) over the ensemble of the turbulent velocity field and subtract the obtained averaged equation from equation (1). This yields equation for the turbulent component q of particles number density

$$\frac{\partial q}{\partial t} - D\Delta q = -\vec{\nabla}\cdot(N\mathbf{u}+\mathbf{Q}) \qquad (15)$$

where $n_p = N + q$, $\mathbf{Q} = \mathbf{u}q - \langle \mathbf{u}q \rangle$. Equation (15) is written in a frame moving with the mean velocity $\mathbf{V}$. The magnitude of $\partial q/\partial t - D\Delta q + \vec{\nabla}\cdot\mathbf{Q}$ can be estimated as q/τ, where τ is the turnover time of turbulent eddies. Thus the turbulent field q is of the order of

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

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

$$\mathbf{J}_T \sim -N\langle \tau\mathbf{u}(\vec{\nabla}\cdot\mathbf{u})\rangle - \langle \tau\mathbf{u}u_j\rangle \nabla_j N. \qquad (16)$$

Using equations (5), (8) and (9) we can reduce the turbulent flux of particles equation (16) to equation (11). Note that the turbulent flux of particles can be estimated also by means of simple dimensional analysis (see Colegrove *et al.*, 1966).

The equilibrium solution of equation (4) is given by

$$(D + D_T)\vec{\nabla}N_0 = N_0\left(D_T\frac{\vec{\nabla}\rho}{\rho} + D\frac{m_p\mathbf{g}}{kT}\right). \qquad (17)$$

Let the vectors $\mathbf{g}$ and $\vec{\nabla}\rho$ be directed along the Z axis. Equation (17) can be rewritten as

$$(3 + Pe)\vec{\nabla}N_0 = -N_0\left(3\frac{m_p}{m_\mu}\Lambda_P^{-1} + Pe\,\Lambda_\rho^{-1}\right) \qquad (18)$$

where

$$\Lambda_P^{-1} = \frac{|\vec{\nabla}P|}{P} = \frac{m_\mu g}{\kappa T}, Pe = \frac{u_0 l_0}{D} = \frac{D_T}{3D}, D_T = \frac{u_0 l_0}{3}$$

P is the fluid pressure, Pe is the Peclet number, D_T is the coefficient of turbulent diffusion, l_0 is the maximum scale of turbulent motions, and u_0 is the typical velocity in scale l_0. The solution of equation (18) has the form

$$N_0 = N_* \exp(-Z/\Lambda_N) \qquad (19)$$

where

$$\Lambda_N = \Lambda_\rho(3 - Pe)\left(3\frac{m_p\Lambda_\rho}{m_\mu\Lambda_P} + Pe\right)^{-1}$$

is the height length scale of the mean particle concentration distribution. Without turbulence (when $Pe < 1$) the height length scale Λ_N is given by

$$\Lambda_N = \Lambda_P\frac{m_\mu}{m_p} \ll \Lambda_\rho$$

where $\Lambda_P \simeq \Lambda_\rho$. In this case all aerosol particles are located in the vicinity of the surface of the planet.

However, in a turbulent atmosphere ($Pe \ll 1$) the situation is drastically changed, i.e. the height length scale of the mean particle concentration is

$$\Lambda_N = \Lambda_\rho\left(1 + 3\frac{m_p\Lambda_\rho}{m_\mu\Lambda_P\,Pe}\right)^{-1}. \qquad (20)$$

In order to analyze equation (20) we use parameters typical for the atmosphere of the Earth. We find from equation (20) that aerosol particles with size less than $1\,\mu$m have a height length scale which is of the order of the density stratification length (Λ_ρ) of the surrounding fluid. Note, in passing, that the latter conclusion explains the occurrence of the small aerosol particles in the upper atmosphere. Thus, turbulent diffusion considerably enlarges the characteristic scale height of the particle distribution.

3. Physics of the effect

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

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

$$\frac{\partial n_p^2}{\partial t} + (\vec{\nabla}\cdot\mathbf{A}) = -n_p^2(\vec{\nabla}\cdot\mathbf{U}) - 2D(\vec{\nabla}n_p)^2 \qquad (21)$$

where $\mathbf{A} = n_p^2\mathbf{U} - D\vec{\nabla}n_p^2$. Consider the evolution of the number density of the particles in a volume V_* in the Lagrangian frame. Integrating equation (21) over the volume V_* we obtain

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

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

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

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

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

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

4. Formation of large-scale inhomogeneities in spatial distribution of particles concentration

Now we study the dynamics of the large-scale distribution of the particle concentration in the small-scale stratified turbulent fluid flow with $\vec{\nabla}\cdot\mathbf{u} = -\mathbf{u}\cdot(\vec{\nabla}\rho/\rho) \neq 0$. Equation (4) for the mean number density of the particles can be rewritten in the form

$$\frac{\partial N}{\partial t} + \vec{\nabla}\cdot[(\mathbf{v}_s - \vec{\lambda}D_T F_0(z))N] = \vec{\nabla}\cdot[(D + D_T F_0(z))\vec{\nabla}N] \tag{22}$$

where we take into account that for the stratified turbulent flow

$$D_{mn} = D\delta_{mn} + D_T F_0(\mathbf{z})\delta_{mn} \tag{23}$$

$$\mathbf{V}_{\text{eff}} = D_T F_0(\mathbf{z})\vec{\lambda}, \quad D_T = \frac{u_0^2\tau_0}{3} \tag{24}$$

(see Elperin *et al.*, 1995). Here we use the notation $\langle\mathbf{u}^2\rangle = u_0^2 F_0(Z)$. Equation (22) has the equilibrium solution (17). Hereafter, we consider the case $Pe \gg 1$, i.e. $D_T \gg D$. Now we study the stability of this equilibrium solution.

We seek a solution of equation (22) of the form

$$N(t,\mathbf{r}) = N_0(\mathbf{r}) + N(t,Z)\exp(i\mathbf{k}\cdot\mathbf{r}_\perp) \tag{25}$$

where the wave vector $\mathbf{k}$ is perpendicular to the axis Z. Substituting equation (25) into equation (22) yields

$$\frac{\partial N}{\partial t} = \frac{1}{m_0}\frac{\partial^2 N}{\partial Z^2} + \mu_0\frac{\partial N}{\partial Z} - \frac{\kappa_0}{m_0}N \tag{26}$$

where

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

Hereafter, we consider the case $F_0(Z) \gg Pe^{-1}$ for all Z. Equation (26) is written in dimensionless form, coordinate Z is measured in units Λ_T, time t is measured in units Λ_T^2/D_T, the wave number k and value λ are measured in units Λ_T^{-1} and $z = \Lambda_T Z$, $v_0 = v_s\Lambda_T/D_T \simeq 3m_p/(m_\mu\,\text{Pe})$, and Λ_T is the characteristic scale of the spatial temperature distribution, the temperature T is measured in units of temperature difference δT in the scale Λ_T, and concentration N is measured in units N_*. The vector $\vec{\lambda} = \lambda\mathbf{e}_z$, $\mathbf{e}_z$ is a unit vector directed along the axis Z.

Substituting

$$N(t,Z) = \Psi_0(Z)\exp(\gamma_0 t)\exp\left[-\frac{1}{2}\int\chi_0\,dZ\right] \tag{27}$$

reduces equation (26) to the eigenvalue problem for the Schrödinger equation

$$\frac{1}{m_0}\Psi_0{}'' + [W_0 - U_0]\Psi_0 = 0 \tag{28}$$

where $W_0 = -\gamma_0$ and the potential U_0 is given by

$$U_0 = \frac{1}{m_0}\left(\frac{\chi_0^2}{4} + \frac{\chi_0{}'}{2} + \kappa_0\right), \quad \chi_0 = \mu_0 m_0 = \frac{F_0{}'}{F_0} + \lambda + \frac{v_0}{F_0}. \tag{29}$$

Now we use a quantum mechanics analogy for the analysis of the formation of inhomogeneities in a spatial distribution of the particle concentration. The instability ($\gamma_0 > 0$) can be excited if there is a region of potential well where $U_0 < 0$. A positive value of W_0 corresponds to the turbulent diffusion, whereas a negative value of W_0 results in the excitation of the instability. Now we introduce a function $f = \ln\langle\mathbf{u}^2\rangle$ and $f' = F_0{}'/F_0$. The potential U_0 can be rewritten as

$$U_0 = \frac{1}{4m_0}[(f' - \lambda)^2 + [\lambda + v_0\exp(-f)]^2 + 4k^2 + 2f'' - 2\lambda' - \lambda^2]. \tag{30}$$

4.1. *Estimation of growth rate of the instability*

In order to estimate the first energy level W_0 we use a modified variational method (e.g. a modified Ritz method). The modification of the regular variational method is required since equation (28) can be regarded as the Schrödinger equation with a variable mass $m_0(Z)$. Now we rewrite equation (28) in a form

$$\hat{H}\Psi_0 = W_0\Psi_0, \quad \hat{H} = U_0 - \frac{1}{m_0}\frac{d^2}{dZ^2}. \tag{31}$$

The modified variational method employs an inequality

$$W_0 \leqslant I, \quad I = \int m_0 \Psi^* \hat{H} \Psi \, dZ \tag{32}$$

where Ψ is an arbitrary function that satisfies a normalization condition

$$\int m_0 \Psi^* \Psi \, dZ = 1. \tag{33}$$

The inequality (32) can be proven if one uses the expanon

$$\Psi = \sum_{p=0}^{\infty} a_p \Psi_0^{(p)},$$

where

$$\sum_{p=0}^{\infty} |a_p^2| = 1 \quad \text{and} \quad \int m_0 (\Psi_0^{(p)})^* \Psi_0^{(k)} \, dZ = \delta_{pk}.$$

The eigenfunctions $\Psi_0^{(p)}$ satisfy the equation $\hat{H}\Psi_0^{(p)} = W_p \Psi_0^{(p)}$. We chose the function Ψ in the form

$$\Psi = A \exp[-\alpha(Z-Z_0)^2/2], \tag{34}$$

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

where the unknown parameters α and Z_0 may be found from the condition of minimum of the function $I(\alpha, Z_0)$ (see equation (32)). Here we use the following spatial distributions of $f(Z)$ and $\lambda(Z)$:

$$f(Z) = b_0 Z^2 \exp(-\beta_0 Z^2) \tag{35}$$

$$\lambda(Z) = (Z - \lambda_0) \exp(-\epsilon_0 Z^2) \tag{36}$$

where $\beta_0 \ll 1$ and $\epsilon_0 \ll 1$. Substituting equation (34) and equation (36) into equation (32) yields

$$I = \frac{(\alpha+b_0)^{1/2}}{2\alpha^{3/2}}\left[2\alpha\left(Z_0 b - \frac{\lambda_0}{2}\right)^2 + (b-\alpha)^2 + 2\alpha k^2\right]$$
$$\times \exp\left(\frac{\alpha b_0 Z_0^2}{\alpha + b_0}\right) - \frac{v_0 \lambda_0}{2} + \frac{v_0 \alpha Z_0}{2(\alpha + b_0)} + \frac{v_0^2}{4}\left(\frac{\alpha + b_0}{\alpha + 2b_0}\right)^{1/2}$$
$$\times \exp\left(-\frac{\alpha^2 b_0 Z_0^2}{(\alpha + b_0)(\alpha + 2b_0)}\right) \tag{37}$$

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

Thus, the modified Ritz method allows us to estimate the growth rate of the instability:

$$\gamma_0 = -W_0 \sim \frac{\lambda_0^2 \eta}{2}\left[1 - \frac{\eta}{2\sqrt{1+c_0}} \exp\left(-\frac{c_0 \lambda_0^2 Y^2}{2(1+c_0)}\right)\right]$$
$$- \frac{k^2}{\sqrt{1-c_0}} \exp\left(\frac{c_0 \lambda_0^2 Y^2}{2(1-c_0)}\right) \tag{38}$$

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

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

.Defining a critical value of Y at which $\gamma_0 = 0$, i.e.

$$Y_{cr}^2 = \frac{2(1+c_0)}{c_0 \lambda_0^2} \ln\left(\frac{\eta}{2\sqrt{1+c_0}}\right) \tag{39}$$

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

$$\gamma_0 \sim \frac{\lambda_0^2 \eta}{2}\left[1 - \left(\frac{4(1+c_0)}{\eta^2}\right)^p\right] \tag{40}$$

where

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

Here we consider a case of $k \ll 1$ which implies long-wave perturbations in the horizontal plane. Note that only this case is important in planetary atmosphere applications. It is seen from equation (39) and equation (40) that the instability is excited when

$$\frac{v_0}{\lambda_0} > 2\sqrt{1+2b_0} \tag{41}$$

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

$$\gamma_0 \sim b_0 \lambda_0^4 Y_0^2 \tag{42}$$

where $Y_0 = \eta - 1 + \sqrt{\eta(\eta-2)}$. It follows from equation (42) that in homogeneous turbulence the instability is not excited. Thus, it is shown here that the equilibrium distribution (19) of particle concentration is unstable. The instability results in the formation of an inhomogeneous distribution of particle concentration. The exponential growth during linear stage of the instability is saturated by nonlinear effects (e.g. two-way coupling of particles and turbulent fluid flow, a change of temperature distribution in the vicinity of temperature inversion layer).

4.2. *Numerical study of the instability*

Equation (28) was solved numerically with turbulent kinetic energy and mean temperature profiles given by equation (35) and equation (36). The extremum of turbulent kinetic energy is located at $Z = 0$, temperature minimum is located at $Z = \lambda_0$ (see equation (35) and equation (36)) and $Z = -H$ is a location of a impermeable for particles boundary (surface of a planet). The boundary condition is determined by integrating equation (22) which yields the condition for Ψ_0

$$\frac{d\Psi_0}{dZ} = -\frac{1}{2}[\chi_0 - 2f']\Psi_0 \quad \text{at } Z = -H. \tag{43}$$

Equation (43) provides zero flux of particles through a horizontal boundary plane $Z = -H$. The second boundary condition is $\Psi_0(Z = \infty) = 0$.

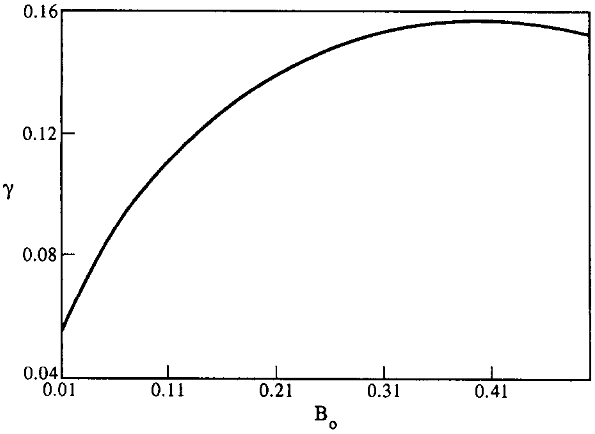

Fig. 1. Dependence of the growth rate of the instability versus b_0 for $\lambda_0 = 1$, $v_0 = 2.2$, $H = 7$

As an example, Fig. 1 shows the dependence of the growth rate of the instability versus b_0 for $\lambda_0 = 1$, $v_0 = 2.2$ and $k \ll 1$. These values of parameters satisfy the necessary condition (41) for the excitation of the instability. The minimum of the function $f(Z)$ describing the spatial distribution of the turbulent kinetic energy is chosen to be located at the height $H = 7\Lambda_T$ from the surface of the planet. These numerical results are in a good agreement with the analytical estimates obtained by means of the modified Ritz method (see Section 4.1). Instability is excited when $0 < b_0 < 0.57$. Note that the analytical estimate obtained by means of the modified Ritz method yields the upper limit for $b_0 = 0.5$.

5. Discussion

Aerosols are produced by the photochemical process in the upper atmosphere of Titan and move with the terminal fall velocity to the lower atmosphere where they are accumulated in the vicinity of the temperature minimum due to effects of turbulent barodiffusion and turbulent thermal diffusion. This causes an increase in the optical depth of the atmosphere. Now we assume that the pressure and density in the Titan atmosphere are related by an adiabatic law: $P\rho^{-\gamma} = \text{const}$. Although the lapse rate in Titan's atmosphere is subadiabatic, the data in Lindal *et al.* (1983) can be fit sufficiently well by this assumption, that it is a good approximation for the atmosphere below the temperature inversion. Equation (18) which describes equilibrium distribution of particles is reduced to

$$\frac{\vec{\nabla} N_0}{N_0} = -\sigma \frac{\vec{\nabla} T}{T} \tag{44}$$

where γ is the specific heats ratio and

$$\sigma = \frac{3(m_p/m_\mu)\gamma + Pe}{(\gamma - 1)(3 + Pe)}.$$

The solution of equation (44) is given by

$$\frac{N_0(\mathbf{r})}{N_*} = \left(\frac{T(\mathbf{r})}{T_*}\right)^{-\sigma} \tag{45}$$

where N_* and T_* are particle concentration and the temperature at a location $\mathbf{r} = \mathbf{r}_*$. It is seen from equation (45) that even in equilibrium particles are concentrated in the vicinity of the minimum in the temperature distribution. Now we estimate the variation of the optical depth

$$\frac{\delta\tau_a}{\tau_a} \propto \frac{1}{\Lambda_T} \int \left(\frac{N_0}{N_*} - 1\right) dZ \propto \left| \left(1 + \frac{\delta T}{T_*}\right)^{-\sigma} - 1 \right|. \tag{46}$$

where we used the relation $T = T_* + \delta T$.

The temperature distribution in the atmosphere of Titan is as follows (e.g. Atreya, 1981). The surface temperature is about 90 K. The temperature decreases with altitude to a minimum of about 70 K at a height of about 40 km and rises to about 170 K at an altitude of approximately 200 km. In the vicinity of the temperature minimum $\Lambda_T \sim 80$–100 km, and $|\delta T|/T_* \sim 0.5$ and $\sigma \sim (\gamma - 1)^{-1} \sim 2.5$. Thus, for these parameters we determined that the increase of the optical depth caused by the accumulation of particles in the region with minimum temperature is $\delta\tau_a/\tau_a \sim 0.64$.

On the other hand, the equilibrium distribution of particles is unstable under certain conditions (see Section 3). The instability results in the additional growth of the optical depth in the region of the temperature inversion. Nonlinear effects (e.g. effect of aerosol particles on the turbulent fluid flow, redistribution of the mean temperature due to accumulated aerosols) saturate the instability. The nonlinear effects are important when the spatial density of particles $m_p N_0$ is of the order of the density ρ of the surrounding fluid. Since the optical depth enhancement is proportional to the ratio of the gas density to the spatial density of aerosols, a substantial enhancement of the optical depth is expected. This enhancement of the aerosol distribution will have a number of consequences for the Titan atmosphere. In the first place it will affect the reflective properties of the atmosphere and result in a change in the computed geometric albedo for a given aerosol composition. In the second place, by changing the position of the aerosols with respect to the gas, it will affect the observed widths of spectral features in the atmosphere. The implications for models of Titan's aerosol are clear. Finally, since the aerosols influence the heat balance in the atmosphere, this too will have to be recomputed with the expected enhancement taken into account.

It may be argued that the increased spatial density of aerosols will result in a corresponding increase in the coagulation rate. This, in turn, will make the aerosol particles grow faster and fall out faster, thus neutralizing the effect. In fact, according to the models of McKay *et al.* (1989), the aerosol particles in this region are of the order of a few tenths of a micron (similar results were obtained by Podolak and Podolak (1980)). Particles this small are strongly influenced by the convective motions of the gas. Indeed the mechanism we describe derives from this source. In order to fall against the convective eddies, the aerosol particles would have to have radii at least an order of magnitude larger. It is not clear that aerosol growth can proceed quickly enough to cause substantial sedimentation on the relevant time scale.

The characteristic time of excitation of the instability $\tau_{inst} \sim (10 - 100)\Lambda_T^2/D_T \sim 10 - 100$ years, were we used that in the atmosphere of Titan at a height of about 40 km the turbulent diffusion coefficient is of order $D_T \sim 10^4$–

$10^5\,cm^2\,s^{-1}$. It is interesting that Titan's year (29.5 Earth years) is the same order of magnitude as this characteristic excitation time. Seasonal changes in the heating rate could act to change the altitude of the temperature inversion in the atmosphere, which would, in turn, affect the vertical distribution of the aerosols. There will, undoubtedly, be additional effects due to influence of the ambient conditions on the details of the microphysics. Such a mechanism could, therefore, help to explain the observed hemispheric albedo dichotomy (Sromovsky *et al.*, 1981), as well as its seasonal change (Caldwell *et al.*, 1992; Lockwood *et al.*, 1986). Clearly detailed numerical models will be necessary before the importance of this mechanism can be fully assessed. Future studies will try to further quantify these effects, and their influence on Titan's atmosphere.

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References

Akhiezer, A. I. and Petleminskii, S. V. (1981) *Methods of Statistical Physics*. Pergamon Press, Oxford.

Atreya, S. K. (1981) *Atmospheres and Ionospheres of the Outer Planets and their Satellites*. Pergamon Press, Oxford.

Avellaneda, M. and Majda, A. J. (1994) Simple examples with features of renormalization for turbulent transport. *Phil. Trans. R. Soc. Lond. A* **346,** 205.

Bar-Nun, A., Kleinfeld, I. and Ganor, E. (1988) Shape, optical properties of aerosols formed by photolysis of acetylene, ethylene and hydrogen cyanide. *J. Geophys. Res.* **93,** 8383.

Caldwell, J., Cunningham, C. C., Anthony, D., White, N. P., Groth, E. J., Hasan, H., Noll, K., Smith, P. H., Tomasko, M. G. and Weaver, H. A. (1992) Titan: evidence for seasonal change—a comparison of Hubble Space Telescope and Voyager images. *Icarus* **97,** 1.

Colegrove, F. D., Johnson, F. S. and Hanson, W. B. (1966) Atmospheric composition in the lower thermosphere. *J. Geophys. Res.* **71,** 2227.

Elperin, T., Kleeorin, N. and Rogachevskii, I. (1995) Dynamics of passive scalar in compressible turbulent flow: large-scale patterns and small-scale fluctuations. *Phys. Rev. E* **52,** 2617.

Elperin, T., Kleeorin, N. and Rogachevskii, I. (1996) Turbulent thermal diffusion of inertial particles. *Phys. Rev. Lett.* **76,** 224.

Elperin, T., Kleeorin, N. and Rogachevskii, I. (1996) Self-excitation of fluctuations of inertial particles concentration in turbulent fluid flow. *Phys. Rev. Lett.* **77,** 5373.

Elperin, T., Kleeorin, N. and Rogachevskii, I. (1997) Turbulent barodiffusion, turbulent thermal diffusion and large-scale instability in gases. *Phys. Rev. E* **55,** 2713.

Hutzell, W. T., McKay, C. P., Toon, O. B. and Hourdin, F. (1996) Simulations of Titan's brightness by a two-dimensional haze model. *Icarus* **119,** 112.

Kleeorin, N. and Rogachevskii, I. (1994) Nonlinear theory of magnetic fluctuations in random flow: the Hall effect. *Phys. Rev. E* **50,** 493.

Landau, L. D. and Lifshitz, E. M. (1987) *Fluid Dynamics*. Pergamon Press, Oxford.

Lindal, G. F., Wood, G. E., Hotz, H. B., Sweetnam, D. N., Eshleman, V. R. and Tyler, G. L. (1983) The atmosphere of Titan: an analysis of the Voyager 1 radio occultation measurements. *Icarus* **53,** 348.

Lockwood, G. W., Lutz, B. L., Thompson, D. T. and Bun, E. S. (1986) The albedo of Titan. *Astrophys. J.* **303,** 511.

McKay, C. P., Pollack, J. B. and Courtin, R. (1989) The thermal structure of Titan's atmosphere. *Icarus* **80,** 23.

Podolak, M. and Danielson, R. E. (1977) Axel dust on Saturn and Titan. *Icarus* **30,** 479.

Podolak, M. and Podolak, E. (1980) A numerical study of aerosol growth in Titan's atmosphere. *Icarus* **43,** 74.

Rannou, P., Cabane, M., Chassefiere, E., Botet, R., McKay, C. P. and Courtin, R. (1995) Titan's geometric albedo: role of the fractal structure of the aerosols. *Icarus* **118,** 355.

Scattergood, T. W., Lau, E. Y. and Stone, B. M. (1992) Titan's aerosols. I—Laboratory investigations of shapes size distributions, and aggregation of particles produced by UV photolysis of model Titan atmospheres. *Icarus* **99,** 98.

Sromovsky, L. A., Suomi, V. E., Pollack, J. B., Krauss, R. J., Limaye, S. S., Owen, T., Revercomb, H. E. and Sagan, C. (1981) Implications of Titan's north–south brightness asymmetry. *Nature* **292,** 698.

Toon, O. B., McKay, C. P., Griffith, C. and Turco, R. P. (1992) A physical model of Titan's aerosols. *Icarus* **95,** 24.

Zeldovich, Ya. B., Molchanov, S. A., Ruzmaikin, A. A. and Sokoloff, D. D. (1988) Intermittency, diffusion and generation in a nonstationary random medium. *Sov. Sci. Rev. C. Math. Phys.* **7,** 1

Zeldovich, Ya. B., Molchanov, S. A., Ruzmaikin, A. A. and Sokoloff, D. D. (1990) *The Almighty Chance*. Word Scientific, London.