TRANSFER EQUATIONS FOR CONSERVATIVE OPTICAL CHARACTERISTICS OF THE ATMOSPHERE

G.O. Zadde and A.V. Podanev

Institute of Atmospheric Optics.
Siberian Branch of the Academy of Sciences of the USSR, Tomsk

Received July 18, 1989

The transfer equations for the absorbing and scattering coefficients and the optical thickness of the molecular and aerosol fractions are derived based on the dynamics of multiphase media. The equations for the molecular optical characteristics are derived for a class of problems in which the temperature and pressure dependence of the coefficients can be neglected. A physical analysis of the equations obtained is given.

It is well known that the time and spatial dependence of the molecular and aerosol absorption and scattering coefficients as well as the corresponding optical thicknesses are completely determined by the dynamics of the gas and aerosol mixture, which is described by the conservation equations, derived in the theory of multiphase media, for the components of the mixture.[1]

The dynamics of a particular spectral optical characteristic is determined by the motion of the components that are optically active for it and for this reason it can exhibit special behavior not characteristic of the dynamics of separate components of the atmosphere. The study of this behavior is important for establishing the relations between the spreading of atmospheric pollutants and the dynamics of the optical indicator.

The traditional approach to the simulation of the dynamics of an optical characteristic is to add to the working mathematical model, after calculating the equations of motion of the gaseous and aerosol components, a block in which the optical characteristics are calculated (typically, this is a multidimensional integral). In this paper we propose to break this chain, i.e., we shall replace in the mathematical model the block describing the dynamics of the gases and aerosols by a block describing the dynamics of the optical characteristics. The purpose of this paper is to derive the corresponding conservation equations for a number of optical characteristics from the laws of conservation of mass of the molecular and aerosol components.

The volume coefficients of the molecular and aerosol scattering ($\sigma_{\lambda mi}$, $\sigma_{\lambda ai}$) and absorption ($\kappa_{\lambda mi}$, $\kappa_{\lambda ai}$) are related through the corresponding average optical characteristics (the superscript "0") with the densities of the components by the relations

$$\varphi_{\lambda b} = \varphi^{0}_{\lambda b}\rho_{b} = \sum_{i=1}^{N_b} \varphi^{0}_{\lambda bi}\rho_{bi} = \sum_{i=1}^{N_b} \varphi_{\lambda}b_{i}, \tag{1}$$

where $\varphi_{\lambda b}$ will be $\sigma_{\lambda m}$ and $\kappa_{\lambda m}$ with $b = m$ and $\sigma_{\lambda a}$ and $\kappa_{\lambda a}$ with $b = a$, and the index "m" corresponds to the molecular component; the index "a" corresponds to the aerosol component; ρ_{mi} is the density of the ith gaseous component; ρ_{ai} is the density of the ith aerosol component, $\rho_m = \sum_{i=1}^{N_m}\rho_{mi}$ is the density of the gas mixture, and $\rho_a = \sum_{i=1}^{N_a}\rho_{ai}$ is the density of the aerosol mixture.

The average molecular scattering coefficient is virtually constant. The average absorption coefficient of the molecular component of an individual line, described by a Lorentzian contour, depends on the pressure and temperature. The degree of the dependence can be estimated from the expression for the line center[2]

$$\chi^{0}_{\lambda mi} = \frac{S_0}{\pi\gamma_0 p}\left[\frac{T_0}{T}\right]^{n}\exp\left\{-1.44\,\frac{E}{T_0}\left[\frac{T_0}{T} - 1\right]\right\}, \tag{2}$$

where $T = 296$ K; S_0, γ_0 and π are constants; E is the energy of the level (300–400 for a strong band and 1500 for a weak band); p is the pressure; and, $n = 1.5$–2.

The position of the extremum for Eq. (2) is determined by the value $T_{*} = \dfrac{E\cdot 1{,}44}{n}$. Let T_{av} be some characteristic temperature (for example, the average daily temperature), when the deviation of $\Delta\kappa^{0}_{\lambda mi}$ from $(\kappa^{0}_{\lambda mi})_{av}$ depending on the deviation of the temperature ΔT from the average temperature, will be

$$\frac{\Delta\chi^{0}_{\lambda mi}}{\left(\chi^{0}_{\lambda mi}\right)_{av}} = n\left[\frac{T_{av} - T_{\bullet}}{T_{av}}\,\frac{\Delta T}{T_{av}}\right].$$

Therefore the relative change in $\kappa^0_{\lambda mi}$ will be small in the case of relatively small $(T_{av} - T_*)$ and ΔT.

Thus it is possible to separate a class of problems in which the average extinction coefficients can be assumed to be virtually constant. Such problems, in particular, can include the problem of the spreading of industrial waste gases with small pressure and temperature gradients (this is observed sufficiently far away from the source).

In what follows we shall assume that $\sigma^0_{\lambda mi}$ and $\kappa^0_{\lambda mi}$ do not depend on the pressure, density, and temperature. This assumption makes it possible to transform by means of a simple linear transformation from the laws of conservation of gaseous components, presented in Ref. 1, to the laws of conservation corresponding to the molecular optical indicator. In this case this equation for the gas and aerosol mixture have the form

$$\frac{d\varphi_{mi}}{dt} + \varphi_{mi} \operatorname{div} \upsilon_m = \sum_{j=1}^{N_m} \Phi^{mm}_{ji} + \sum_{j=1}^{N_a} \Phi^{am}_{ji} - \operatorname{div}(\varphi_{mi} w_{mi}), \tag{3}$$

where

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \upsilon_m \cdot \nabla = \frac{\partial}{\partial t} + \upsilon_x \frac{\partial}{\partial x} + \upsilon_y \frac{\partial}{\partial y} + \upsilon_z \frac{\partial}{\partial z} ;$$

υ_m is the weighted-mean velocity of the gaseous mixture; $\varphi_{m1} = (\sigma_{\lambda m1}, \kappa_{\lambda m1}, \alpha_{\lambda m1})$ $\alpha_{\lambda m1}$ the extinction coefficient of the ith gaseous component; $\Phi^{mm}_{j1} = J^{mm}_{j1} \varphi^0_{m1}$ is the intensity of the change in the ith molecular optical characteristic owing to the gas-phase reaction of the transition of the jth molecular component into the ith molecular component; Φ^{am}_{j1} is the same for the reaction of the transition of the jth aerosol fraction into the ith molecular fraction; and, $w_{mi} = (\upsilon_{mi} - \upsilon_m)$ is the rate of diffusion of the ith component.

We note that if $\sum\limits_{i=1}^{N_m} \sum\limits_{j=1}^{N_m} J^{mm}_{j1} = 0$ by virtue of the law of conservation of mass, then

$$\sum_{i=1}^{N_m} \sum_{j=1}^{N_m} \Phi^{mm}_{ji} = \sum_{i=1}^{N_m} \sum_{j=1}^{N_m} J^{mm}_{ji} \varphi^0_{mi} =$$

$$= \sum_{j=1}^{N_m} \sum_{i=j}^{N_m} J^{mm}_{ji} \left(\varphi^0_{mj} - \varphi^0_{mi}\right) \neq 0,$$

i.e., in the general case the sum of the sources of the optical characteristics is not equal to zero.

We also note that it does not follow from the fact that $\sigma^0_{\lambda mi}$ is constant that $\sigma^0_{\lambda m}$ is constant because it depends on the densities of the components. For this reason, for the total volume molecular optical indicator $\varphi_m = (\sigma_{\lambda m}, \kappa_{\lambda m}, \alpha_{\lambda m})$ we shall have

$$\frac{d\varphi_m}{dt} + \varphi_m \operatorname{div} \upsilon_m = \sum_{j=1}^{N_m} \sum_{i=j}^{N_m} J^{mm}_{ji} \left(\varphi^0_{mj} - \varphi^0_{mi}\right) +$$

$$+ \sum_{i=1}^{N_m} \sum_{j=1}^{N_a} \Phi^{am}_{ji} - \operatorname{div} \sum_{i=1}^{N_m} \varphi_{mi} w_{mi} . \tag{4}$$

We shall present later a physical interpretation of the terms of this equation which is identical to the physical Interpretation for the more general equation (8).

The average spectral aerosol interaction constant $\varphi^0_{\lambda a} = (\sigma^0_{\lambda a}, \kappa^0_{\lambda a}, \alpha^0_{\lambda a})$ for uniform particles depends on the radius r and the complex index of refraction m

$$\varphi^0_{\lambda a} = 3/(4\rho^*_{ai}) K(\mu, m), \ \mathrm{m}^2/\mathrm{kg}, \tag{5}$$

where ρ^*_{a1} is the characteristic density of the ith aerosol substance; K is the interaction factor (the effective cross section); and, $\mu = 2\pi r/\lambda$ is the interaction parameter.

Therefore if for the ith aerosol component we choose a collection of particles of the same size (r) and composition (ρ^*_{a1}, m) then from the law of conservation of this aerosol component ρ_{ai} (Ref. 1) it is easy to derive a corresponding evolutionary equation for the spectral volume optical interaction coefficient:

$$\frac{d\varphi_{ai}}{dt} + \varphi_{ai} \operatorname{div} \upsilon_m = \sum_{j=1}^{N_m} \Phi^{ma}_{ji} + \sum_{j=1}^{N_a} \Phi^{aa}_{ji} -$$

$$- \operatorname{div}\left(\varphi^0_{ai} w_{am}\right) - \operatorname{div}\left(\varphi_{ai} w_{ai}\right) . \tag{6}$$

where $\varphi_{ai} = (\sigma_{\lambda ai}, \kappa_{\lambda ai}, \alpha_{\lambda ai})$ $\alpha_{\lambda ai}$ is the extinction coefficient of the ith aerosol component; $\Phi^{ma}_{ji} = J^{ma}_{ji} \varphi^0_m$ is the intensity of the change in the ith aerosol optical characteristic owing to the gas and aerosol reaction of the transition of the jth molecular component into the ith aerosol component; J^{ma}_{ji} is the average rate of the corresponding reaction; Φ^{aa}_{ji} is the same for the reaction of the transition of the jth aerosol fraction into the ith aerosol fraction; $w_{am} = (\upsilon_a - \upsilon_m)$ is the diffusion velocity of the aerosol mixture as a whole relative to the average velocity of the gas; $w_{ai} = (\upsilon_{ai} - \upsilon_a)$ is the diffusion velocity of the ith aerosol fraction relative to the center of mass of the aerosol mixture.

The equation corresponding to Eq. (4) for the aggregate volume aerosol optical Interaction coefficient will have the form

$$\frac{d\varphi_a}{dt} + \varphi_a \operatorname{div}\upsilon_m = \sum_{i=1}^{N_a}\sum_{j=1}^{N_m} \Phi_{ji}^{ma} + \sum_{j=1}^{N_a}\sum_{i=j}^{N_a} J_{ji}^{aa}\left(\varphi_{aj}^0 - \varphi_{ai}^0\right) - \operatorname{div}\left(\varphi_a w_{am}\right) - \operatorname{div}\sum_{i=1}^{N_a}\varphi_{ai}w_{ai}, \qquad (7)$$

where

$$\varphi_{ai} = \left(\sigma_{\lambda ai},\ \chi_{\lambda ai},\ \alpha_{\lambda ai} = \sigma_{\lambda ai} + \chi_{\lambda ai}\right).$$

$$\varphi_a = \left(\sigma_{\lambda a},\ \chi_{\lambda a},\ \alpha_{\lambda a}\right)$$

For the total molecular-aerosol optical characteristic we obtain from Eqs. (4) and (7) the following evolutionary equation

$$\frac{\partial\varphi}{\partial t} + \operatorname{div}\left(\varphi\cdot\upsilon_m\right) = \sum_{j=1}^{N_m}\sum_{i=j}^{N_m} J_{ji}^{mm}\left(\varphi_{mj}^0 - \varphi_{mi}^0\right) + \sum_{i=1}^{N_m}\sum_{j=1}^{N_a} J_{ij}^{ma}\left(\varphi_{aj}^0 - \varphi_{mi}^0\right) + \sum_{j=1}^{N_a}\sum_{i=j}^{N_a} J_{ji}^{aa}\left(\varphi_{aj}^0 - \varphi_{ai}^0\right) - \operatorname{div}\left[\sum_{i=1}^{N_m}\varphi_{mi}w_{mi} + \varphi_a w_{am} + \sum_{i=1}^{N_a}\varphi_{ai}w_{ai}\right], \qquad (8)$$

where $\varphi = (\sigma_{\lambda m} + \sigma_{\lambda a},\ \kappa_{\lambda m} + \kappa_{\lambda a},\ \alpha_{\lambda m} + \alpha_{\lambda a})$.

This balance relation shows that the time dependence of the absorption, scattering, and extinction coefficients at a given point in space accompanying the motion of the gas and aerosol mixture occurs as a result of convective-advective displacement of the medium (second term), gas-phase chemical reactions (third term), gas-aerosol interphase transitions (fourth term), aerosol-aerosol interactions and transitions (fifth term), and three types of diffusion: diffusion of molecular components, diffusion of the aerosol as a whole relative to the gas mixture, and motion of the aerosol fractions relative to the motion of the center of mass of the aerosol.

Since we have in mind applications to environmental protection the problems, we shall orient the *OZ*-axis of the Cartesian coordinate system perpendicular to the earth's surface and we shall introduce the vertical optical thicknesses

$$\tau_{\lambda mz} = \int_0^z \alpha_{\lambda m}dz\ ; \qquad \tau_{\lambda az} = \int_0^z \alpha_{\lambda a}dz \qquad (9)$$

To obtain the corresponding evolutionary equations for the optical thicknesses, we shall integrate from 0 to z the equations describing the spreading of the molecular (4) and aerosol (7) optical indicators. Then from Eq. (4) we shall have for $\tau_{\lambda mz}$

$$\frac{\partial\tau_{\lambda mz}}{\partial t} + \left(\alpha_{\lambda m}\upsilon_z\right)\Big|_{z=z} - \left(\alpha_{\lambda m}\upsilon_z\right)\Big|_{z=0} + \int_0^z\left[\frac{\partial}{\partial x}\left(\alpha_{\lambda m}\upsilon_y\right) - \frac{\partial}{\partial y}\left(\alpha_{\lambda m}\upsilon_y\right)\right]dz =$$

$$= \sum_{j=1}^{N_m}\sum_{i=j}^{N_m}\int_0^z J_{ji}^{mm}\left(\alpha_{\lambda mj}^0 - \alpha_{\lambda mi}^0\right)dz + \sum_{i=1}^{N_m}\sum_{j=1}^{N_a}\int_0^z J_{ji}^{am}\alpha_{\lambda mi}^0\,dz - \left[\sum_{i=1}^{N_m}\alpha_{\lambda mi}w_{miz}\right]\Bigg|_{z=z} + \left[\sum_{i=1}^{N_m}\alpha_{\lambda mi}w_{miz}\right]\Bigg|_{z=0} - \int_0^z\frac{\partial}{\partial x}\left[\sum_{i=1}^{N_m}\alpha_{\lambda mi}w_{mix}\right]dz - \int_0^z\frac{\partial}{\partial y}\left[\sum_{i=1}^{N_m}\alpha_{\lambda mi}w_{miy}\right]dz. \qquad (10)$$

Taking into account the fact that

$$\int_0^z \alpha_{\lambda m}\upsilon dz = \int_0^z \upsilon\left[\frac{\partial}{dz}\int_0^z\alpha_{\lambda m}dz\right]dz = \int_0^z \upsilon d\tau_{\lambda mz} = \upsilon\tau_{\lambda mz}\Big|_{z=z} - \upsilon\tau_{\lambda mz}\Big|_{z=0} - \int_0^z\tau_{\lambda mz}\frac{\partial\upsilon}{\partial z}dz, \qquad (11)$$

and

$$\tau_{\lambda mz}\Big|_{z=0} = 0,$$

we can put the fourth term in Eq. (10) into the following form

$$\int_0^z\left[\frac{\partial}{\partial x}\left(\alpha_{\lambda m}\upsilon_x\right) + \frac{\partial}{\partial y}\left(\alpha_{\lambda m}\upsilon_y\right)\right]dy =$$

$$= \frac{\partial}{\partial x}\left[\left(v_x\tau_{\lambda mz}\right)\Big|_{z=z} - \left(v_x\tau_{\lambda mz}\right)\Big|_{z=0} - \int_0^z \tau_{\lambda mz}\frac{\partial v_x}{\partial z}\,dz\right] + \frac{\partial}{\partial y}\left[\left(v_y\tau_{\lambda mz}\right)\Big|_{z=z} - \left(v_y\tau_{\lambda mz}\right)\Big|_{z=0} - \int_0^z \tau_{\lambda mz}\frac{\partial v_y}{\partial z}\,dz\right]. \tag{12}$$

The evolutionary equation for the vertical aerosol optical thickness is derived analogously — by integrating Eq. (7)

$$\frac{\partial\tau_{\lambda az}}{\partial t} + \left(\alpha_{\lambda a}v_z\right)\Big|_{z=z} - \left(\alpha_{\lambda a}v_z\right)\Big|_{z=0} + \frac{\partial}{\partial x}\left[\left(v_x\tau_{\lambda az}\right)\Big|_{z=z} - \int_0^z \tau_{\lambda az}\frac{\partial v_x}{\partial z}\,dz\right] + \frac{\partial}{\partial y}\left[\left(v_y\tau_{\lambda az}\right)\Big|_{z=z} - \int_0^z \tau_{\lambda az}\frac{\partial v_y}{\partial z}\,dz\right] =$$

$$= \sum_{j=1}^{N_a}\sum_{i=j}^{N_a}\int_0^z J^{aa}_{ji}\left(\alpha^0_{\lambda aj} - \alpha^0_{\lambda ai}\right)dz + \sum_{i=1}^{N_m}\sum_{j=1}^{N_a}\int_0^z J^{ma}_{ji}\,\alpha^0_{\lambda ai}\,dz - \left(\alpha_{\lambda a}w_{amz}\right)\Big|_{z=z} + \left(\alpha_{\lambda a}w_{amz}\right)\Big|_{z=0} - \frac{\partial}{\partial x}\left[\left(w_{amx}\tau_{\lambda az}\right)\Big|_{z=z} - \int_0^z \tau_{\lambda az}\frac{\partial w_{amx}}{\partial z}\,dz\right] - \frac{\partial}{\partial y}\left[\left(w_{amy}\tau_{\lambda az}\right)\Big|_{z=z} - \int_0^z \tau_{\lambda az}\frac{\partial w_{amy}}{\partial z}\,dz\right] -$$

$$- \left[\sum_{i=1}^{N_a}\alpha_{\lambda ai}w_{aiz}\right]\Bigg|_{z=z} + \left[\sum_{i=1}^{N_a}\alpha_{\lambda ai}w_{aiz}\right]\Bigg|_{z=0} - \int_0^z \frac{\partial}{\partial x}\left[\sum_{i=1}^{N_a}\alpha_{\lambda ai}w_{aix}\right]dz - \int_0^z \frac{\partial}{\partial y}\left[\sum_{i=1}^{N_a}\alpha_{\lambda ai}w_{aiy}\right]dz. \tag{13}$$

This dynamical equation shows that the time dependence of the optical thickness on a vertical path near the ground from 0 to z (first term) is produced by the vertical efflux of aerosol from the path from the top (second term), injection of aerosol on the path from the surface of the earth (third term), and advective displacement of the optical thickness (4-, 5, 6-, and 7-th terms). The eighth term in Eq. (13) is responsible for the change in the optical thickness owing to all aerosol-aerosol reactions occurring with a change in the extinction coefficient ($\alpha^0_{\lambda aj} - \alpha^0_{\lambda ai}$), and the ninth term corresponds to the change produced by molecular-aerosol transformations.

A remarkable feature of the equations of dynamics for the optical thicknesses (10) and (13) is that their dimension is one less than that of the equations of dynamics of the corresponding optical indicator. This fact will significantly simplify the analytical and numerical analysis of these equations and will therefore make them more useful in practice.

In conclusion we shall point out a number of interesting properties of the evolutionary equation.

1. For $v_z\big|_{z=z} = v_z\big|_{z=0} = 0$ then $\tau_{\lambda az}$ will not depend on the distribution of the vertical velocity of the gas on the path.

2. If $\partial v_x/\partial z = 0$ then v_x is the longitudinal component of the advective velocity of the gas, which will affect the value of $\tau_{\lambda az}$ by simply transporting it. The same conclusion also valid for v_y.

We hope that the new approach, presented above, to the dynamics of the elements of the optical weather, which will make it possible to examine the processes through the prism of the equations of conservation, will turn out to be productive and helpful.

REFERENCES

1. R.I. Nigmatullin, *Dynamics of Multiphase Media* [in Russian], Nauka, Moscow (1987), Part I, 464 pp.

2. Yu.S. Makushkin, A.A. Mitsel, L.I. Nesmelova, et al., "*Automatic system for calculating absorption characteristics of the atmosphere*," VINITI, No. 3685-84, Moscow (1984), Parts I and II.