ON MOISTENING OF ASH PARTICLES IN SMOKE PLUMES OF INDUSTRIAL SOURCES

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Moistening of ash particles occurring in the humid atmosphere is one of the main factors decreasing the accuracy of the lidar measurements of thickness of smoke emissions. Theoretical investigation of the growth of water coating of smoke particles under different meteorological conditions within the zone of emission has been carried out based on the Gaussian model of smoke plume with slant axis and its parameters.

Numerical calculations have shown that in the case of high initial moisture content of the emissions near the source in the smoke plume the zone appears in which water vapor is supersaturated and the effect of particle moistening is significant. Seasonal trends and diurnal variations in temperature and humidity in the surface layer of the atmosphere also substantially affect moistening. Length of the zone of moistening of ash particles is maximum at night in winter under conditions of light breeze.

The possibility of retrieving the initial mass concentration of the dry aerosol in the smoke plume has been shown based on lidar measurements of the scattering coefficient withinin the zone of maximum degree of moistening of smoke plume.target.

1. INTRODUCTION

One of the problems of diagnostics of aerosol plumes of industrial sources is the determination of the mass concentration of aerosol emissions from smoke-stacks. In lidar sensing of smoke plumes this parameter is calculated based on the measurements of the profile of the scattering coefficient in aerosol parcel along the sensing path.[1,2]

Theoretical relationship between the mass concentration M and the scattering coefficient σ_s is given by the formula $M = \mu\sigma_s$, where the proportionality factor μ depends on aerosol microstructure and is calculated, as a rule, based on the model function of particle size distribution.

However, the experimental investigations have shown[1] that under real conditions moistening of ash particles occurring in the humid atmosphere substantially affects the factor μ in addition to the natural variations of aerosol density that can lead to considerable deviations of the factor μ from its theoretical estimates. In this connection it seems to be important to obtain an *a priori* estimate of a possible degree of moistening of solid particles of the smoke emissions and its variation along the sensing path in order to increase the accuracy and reliability of retrieving the factor μ.

Distinguishing features of this problem are the strong variability of the moisture field in the smoke plume depending on the meteorological conditions within the zone of emission and its characteristics. Moreover, the smoke plume by itself can be the source of water vapor injections into the atmosphere. This leads to the appearance of the zones of local supersaturation ($f > 1$, where f is the relative humidity) in the plume and, correspondingly, to sharp growth of water coating of the aerosol particles.

2. MOISTENING OF SOLID PARTICLES IN THE HUMID ATMOSPHERE

The main mechanisms of moistening of aerosol particles are adsorption and condensation of water vapors[3] on their surfaces. For the relative humidity $f < 1$ the equilibrium radius of a moist particle can be determined as[3]

$$a(f) = a_0\left(1 - C\frac{\varepsilon_v}{\ln f}\right)^{1/3};\; C = \frac{\Phi\rho_s M_w}{\rho_w M_s}, \quad (1)$$

where a_0 is the radius of the dry core; ε_v is the volume fraction of the soluble substances in the particle; p_s and M_s are the density and average molar mass of soluble substances, respectively; p_w and M_w are the density and average molar mass of water; and, Φ is the osmotic factor. Formula (1) is written down disregarding the correction for the curvature of drops and is valid for $a_0 \geq 0.1$ μ*m* (see Ref. 3).

As has been mentioned above, the appearance of supersaturation zone in the medium ($f > 1$) in the vicinity of the

$$\frac{da}{dt} = K\frac{s + \Theta(c)}{a + \xi};\frac{\partial F}{\partial t} + \frac{\partial}{\partial a}\left(F\frac{da}{dt}\right) = 0;$$

$$\frac{ds}{dt} = -\frac{Q_e}{R_c T}\frac{d(\ln T)}{dt} - 4\pi KN\frac{\rho_w}{\rho_v^s(T)}\int_0^\infty [s + \Theta(c)]\frac{a^2}{a + \xi}F(a)da. \quad (2)$$

Here $s = f - 1$ is the supersaturation degree, Θ(c) is the function which takes into account reduction of the vapor pressure above the solution having the concentration c, T

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and R_c are the temperature and gas constant of vapor, Q_e is the specific heat of water evaporation, $p_v^s(T)$ is the density of saturated vapor at the temperature T, K and ξ are the coefficients determined in Ref. 4, $F(a)$ is the size distribution function of aerosol particles, and N is the particle number density.

Variation of a complex refractive index $m = n + \text{A}c$ of a solid particle caused by appearance of water coaling around this particle can be approximately taken into account as[1]

$$n(f) = n_w + (n_d - n_w)\,[a(f) / a_0)^{-3} ;$$

$$k(f) = k_w + (k_d - k_w)\,[a(f) / a_0)^{-3} , \qquad (3)$$

where n_w and K_w are for water and n_d and K_d are for dry core (according to the data obtained in Ref. 3 for aerosol particles from industrial regions $n_d \approx 1.45$ and $K_d = 0.01$).

3. DIFFUSE SPREADING OF THE SMOKE PLUME

To calculate the humidity field in the smoke plume $f(x, y, z)$, we make use of the model of the Gaussian plume applicable under assumption of steady-state gas stream[5]

$$\rho_v(x,y,z) = \frac{Q}{2\pi V_w \sigma_y(x)\sigma_z(x)}\exp\left[-\frac{y^2}{2\sigma_y^2}\right]\times$$

$$\times\left(\exp\left[-\frac{(z-H_0)^2}{2\sigma_z^2}\right] + \exp\left[-\frac{(z+H_0)^2}{2\sigma_z^2}\right]\right) + \rho_{v\infty} \qquad (4)$$

and

$$f(x, y, z) = \rho_v(x, y, z) / \rho_v^s\ T(x, y, z)) . \qquad (5)$$

Here $Q = \pi R_0^2 V_0 \rho_v (0, 0, z_s)$ is the strength of a continuous smoke source, V_0 is the rate of gas emission from the mouth of the smoke-stack with the radius R_0, p_v is the density of water vapor inside the plume, $p_{v\infty}$ is the density of water vapor outside it, H_0 is the effective altitude of the source, V_w is the wind speed averaged over the layer $z = (0 - H_0)$, z is the altitude, y is the distance from the plume axis in the transverse direction, x is the distance along the plume, $\sigma_y(x)$ and $\sigma_z(x)$ are the-horizontal and vertical standard deviations for the number density distribution of the impurities in the plume, and z_s is the source altitude. The formula analogous to Eq. (4) can be written down for the temperature T of gaseous medium as well.

Diffuse spreading of aerosol .impurity can be also described within the scope of the Gaussian model of the plume with slant axis[6] taking into account gravitational sedimentation of the particles. In this case the effective altitude of the plume axis H_0 proves to be the function of the distance x

$$H(x) = H_0 - \frac{1}{V_w}\int_0^X V_s(a)\,\mathrm{d}x ,$$

where $V_s(a)$ is the rate of sedimentation of the particle with the radius a. Since the shape of the moist particle is nearly spheroidal, the Stokes formula can be used

$$V_s = 2a^2 g\rho / g\mu_d,$$

where g is the acceleration of gravity, p is the particle density, n_d is the coefficient of dynamic viscosity of air.

Then the relation for the particle number density of the aerosol impurity $N(x,y,z)$ takes the form

$$N(x,y,z) = N_0\frac{R_0^2 V_0}{2V_w\sigma_y\sigma_z}\exp\left[-\frac{y^2}{2\sigma_y^2}\right]\times$$

$$\times\int_0^\infty F(a)\exp\left[-\frac{(z-H(x,a))^2}{2\sigma_z^2}\right]\mathrm{d}a. \qquad (6)$$

In Eq. (6) N_0 is the initial number density of aerosol particles (near the mouth of the source).

Let us discuss the problem of the rate of condensation growth of particles. Using Eq. (2) we can estimate the characteristic time T_C required for the produced vapor excess to be absorbed by the system of N_0 moist particles

$$\tau_c \simeq (4\,\pi\,D_v\,\bar{a}\,N_0\,)^{-1}$$

where $\bar{a}$ is a mean size of moist particles and D_v is the coefficient of vapor diffusion. The time τ_c is to be compared with the characteristic time of variation in the humidity and temperature fields in the smoke plume τ_p which can be estimated from Eq. (4)

$$\tau_c \approx (4\pi D_v \bar{a} N_0)^{-1},$$

where C_i are the virtual diffuse coefficients given in Ref. 5 $\left(C_i = \sqrt{2\frac{\sigma_i}{x}}\right)$. Then, for typical parameters of emissions[2] $V_0 = 20$ m/s, $R_0 = 5$ m, $N_0 \simeq 10^4$ cm^{-3}, and $\bar{a} = 10$ μm, for the wind speed $V_w = 5$ m/s under conditions of neutral stratification of the atmosphere we obtain $\tau_C/\tau_P \le 10^{-3}$. Therefore, the process of condensation of the vapor excess on the particles can be considered instantaneous and the size of moist particles equilibrium.

4. DISCUSSION OF RESULTS

Let us now discuss the results of numerical calculations of moistening of aerosol impurity in the smoke plume from Eqs. (1)–(6).

Figure 1*a* shows the relative humidity f on the plume axis ($y = 0$ and $z = H_0$) for different mass fraction of water vapor in gaseous emissions $\varphi = M_v/M_g$ as a function of the normalized distance x/R_0. Here M_g and M_v are the total mass output of gaseous products of combustion and mass outflow of water vapor per second, respectively. It can be seen from the figure that for high vapor content in outgoing gases of the plume there appears the zone within which the vapor is supersaturated (dashed curves in Fig. 1). Degree of this supersaturation increases with φ. The same pattern can be observed in the case of change in the initial temperature T_0 of the emission (Fig. 1*c*).

Since under real conditions the aerosol particles arc always emitted together with gases, then according to the above estimates these particles will absorb the vapor excess for rather short time τ_C (solid curves in Figs. 1*a*–*c*).

An effect of the ratio of the speed of the outgoing gases V_0 to the average wind speed V_w on the axial profile of humidity in the smoke plume is illustrated by Fig. 1*b*. It can be seen from the figure that the growth of the parameter V_w/V_0 corresponding to the intensification of the atmospheric diffusion under conditions of constant insolation results in shortening the length of the zone of supersaturation in the plume. At the same time the variation of the ratio V_w/V_0 practically has no effect on the maximum supersaturation.

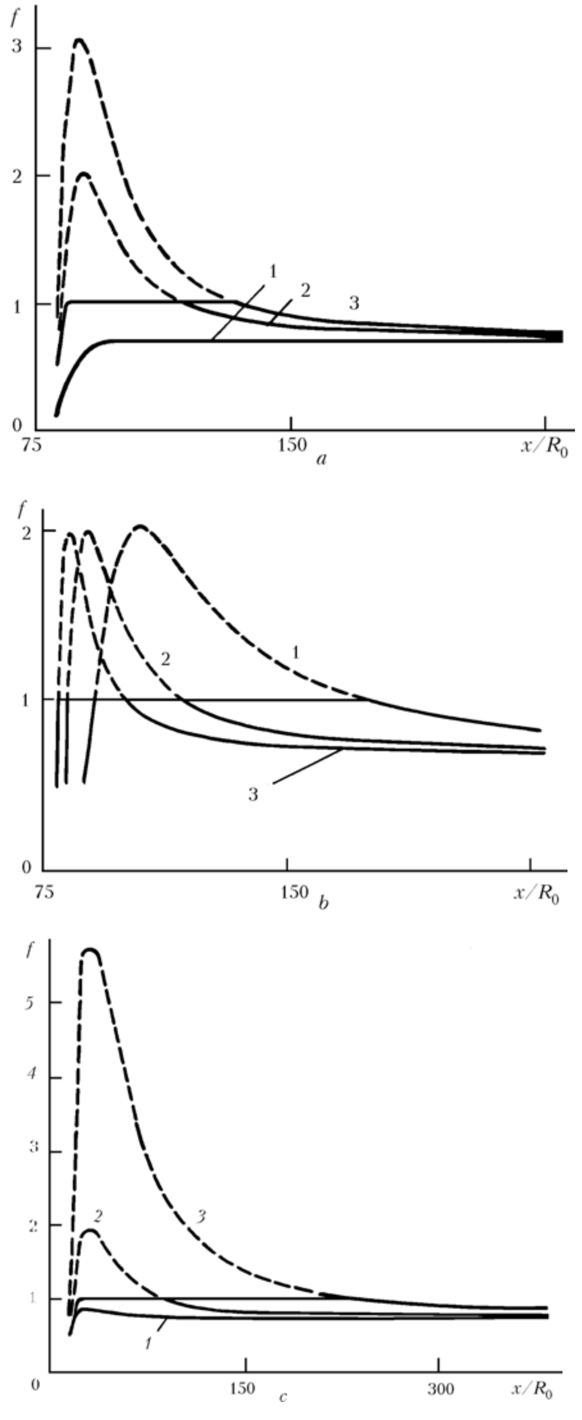

FIG. 1. Relative humidity on the axis of the smoke plume as a function of dimension/ess distance for T_0 = 100°C, z_s = 100 m, f_{oo} = 0.7, *and stable atmosphere. The parameters used in calculation are as follows:* V_w/V_0 = 0.2: *a) for different initial moistening of the jet stream:* φ = 0.1 (1), 0.5 (2), and 0.8 (3). *b) For different ratio* V_w/V_0 = 0.01 *(1),* 0.1 *(2), and* 1.0 *(3),* φ = 0.5. *c) For different initial temperature of the emission* T_0 = 60 *(1);* 100 *(2), and* 150°C *(3).*

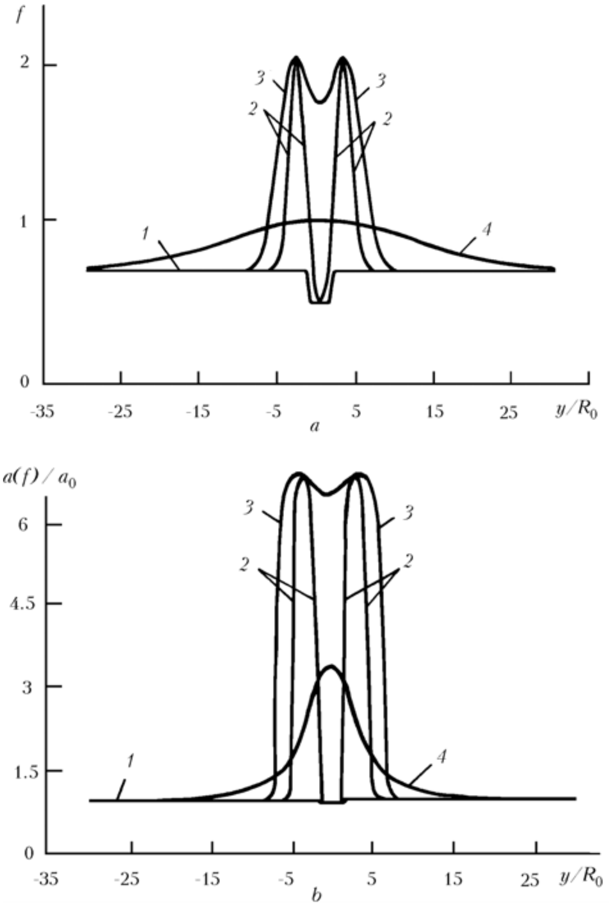

FIG. 2. The profile of relative humidity (a) and the degree of moistening of the particles (b) in the smoke jet stream at different distances from the source x/R_0 = 0 *(1),* 8 *(2),* 15 *(3), and* 45 *(4). The other parameters correspond to those of Fig. 1.*

Figure 2 shows the model calculations of the transverse profile of the relative humidity at different distances from the source disregarding the condensation on the particles. An analysis of the curves shows that near the mouth of the smokestack ($x/R_0 \lesssim 45$) the profile of f substantially differs from the Gaussian one. The dip on the jet stream axis is caused by the fact that turbulent mixing of gases flowing out with air and determining an increase of f occurs in the periphery of the jet stream earlier than in its central parts. Spreading of the plume (curves *3* and *4* in Fig. 2*a)* occurs as the distance from the source increases.

To obtain the dependence of the humidity field on the diurnal variations and the seasonal trends in the temperature and humidity of surrounding air is of great importance for investigation of moistening of aerosol particles in the smoke plume.

For this purpose a series of calculations of the dependence $f(x)$ in summer and winter in the day time and at night (Figs. 3*a* and *b)* was carried out using regional and seasonal models of the atmosphere. The parameters of the emission were taken to be constant and typical of the emissions of the heat-and-power stations: V_w/V_0 = 0.2, φ = 0.1, and the temperature of outgoing gases T_0 was equal to 100°C.

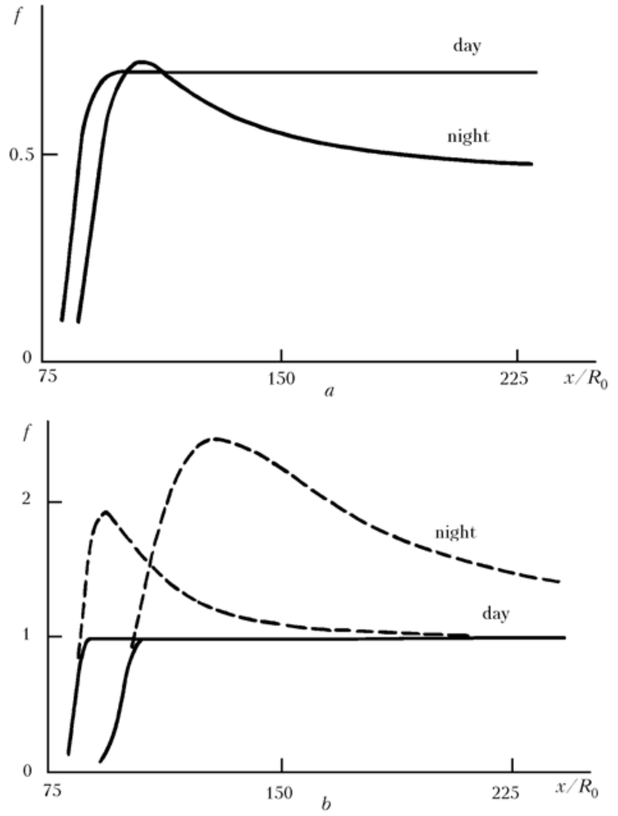

FIG. 3. Variation in the relative humidity on the axis of the smoke jet stream at different distances from the smoke source in summer (a) and winter (b) in the day time and at night.

Calculation has shown that the highest relative humidity in the smoke plume is attained in winter as could be expected (Fig. 3*b*). Diurnal variations in f in the plume depend also on season. If in winter at night the humidity of the plume was much higher than in the day time then in summer these differences became insignificant and were primarily determined by the diurnal behavior of the relative humidity of air f_∞. The shift of the maximum in the function $f(x)$ toward greater x at night can be explained by the fact that at night the surface layer of the atmosphere is, as a rule, more stable[7] and diffusion of the smoke jet stream proceeds slower than in the day time.

According to Eqs. (1)-(3) variation in relative humidity in the smoke plume alters the size and changes the structure of the smoke particles. Calculated dependences of the degree of moistening $a(f)/a_0$ and the refractive index n of the particles in the visible range of wavelengths on dimensionless distance from the source x/R_0 are shown in Figs. 4*a* and *b*. The parameters used for calculation correspond to those of Fig. 3. The initial mass concentration of the solid aerosol M_0 was equal to $1 \cdot 10^{-3}$ kg/m^3 (Ref. 2). The volume fraction of the soluble substances of the particles $\varepsilon_v \simeq 0.06$ (Eq. (1)) was borrowed from Ref. 8 (Θ(c) $\simeq 0$). Small values of ε_v cause very low degree of moistening of the smoke particles of the heat-and-power station for $f < 1$.

The normalized log-normal distribution was used as a particle size distribution function of a volatile fraction of ash

$$F(a) = \frac{1}{\sqrt{2\pi}\ln 10\, a_m} \exp\left[-\frac{\log^2(a_m/a_0)}{2\sigma^2}\right], \qquad (7)$$

which, as was shown in Ref. 9, fits the experimental data quite well. The parameters of distribution, i.e., the mean geometric size of the ash particles a_m and the variance of the logarithms of the particle size were taken as follows: $a_m = 12.5$ /μm; $\sigma = 0.2$ (ash of ball mills[9]).

Degree of moistening of the particles $a(f)/a_0$ in the case of supcrsaturation in the gaseous stream $(s > 0)$ was determined by solving the system of equations (2).

The results shown in Figs. 1–4 indicate that the maximum degree of moistening of the smoke particles is to be expected near the mouth of the source in winter under conditions of light breeze (Fig. 16) for weak atmospheric turbulence. In this case, from the viewpoint of optics, it seems to be very probable that within the zone of high degree of moistening the behavior of smoke particles will be similar to that of water drops (ice crystals) with the radius $a(f)$ $(m \simeq m_w)$, i.e., the effect of nonuniformity of the internal structure of particles can be ignored in calculation of their optical characteristics in the first approximation.[10] This circumstance opens the possibility to retrieve quite simply the sought-after mass concentration of dry aerosol given that the path of sensing of the plume passes through the zone of maximum degree of moistening.

Really, the maximum degree of moistening of the aerosol particles $a(s)/a_0$ in the case of supersaturation in gaseous stream can be determined starting from Eq. (2)

$$s\rho_v^s(T) \simeq \frac{4\pi}{3}N(x,y,z)\rho_w \int_0^\infty F(a;x,y,z)[a^3(s) - a^3(s=0)]\,da. \qquad (8)$$

The term $d(\ln T)/dt$ is omitted in Eq. (8) since the estimates have shown that for real mass concentrations of solid particles in the smoke plume ($M_0 \le 10^{-2}$ kg/m^3, see Refs. 1–2) the effect of liberation of the latent heat of condensation can be ignored.

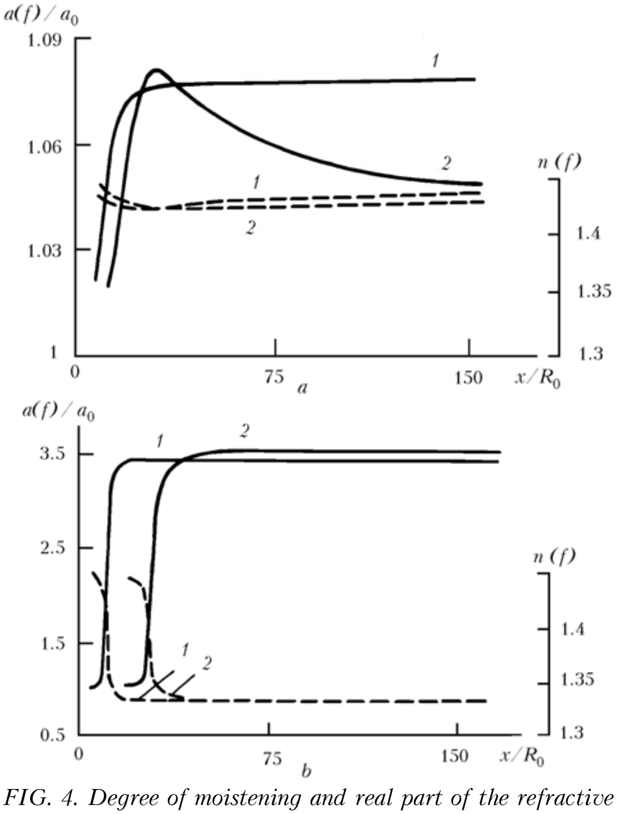

FIG. 4. Degree of moistening and real part of the refractive index of the ash particles in the plume as functions of the dimensionless distance in summer (a) and winter (b) in the day time and at night.

Theoretical investigations of the condensation growth of systems of polydisperse particles[4] indicate that the size distribution function of growing drops gets narrower and shifts toward the larger radii as time goes by. This is a consequence of higher rate of growth of the small drops in comparison with that of the larger drops (see Eq. (2) and Fig. 5). In this connection in Eq. (8) we can approximately assume (that $F(a)\,|_{s>0} \simeq \sigma(a - a_m(s))$ for $s > 0$, where $\sigma(a)$ is the delta function of Dirac and $a_m(s)$ is the maximum size of grown-up particles with the initial radius *a*.

After inverting Eq. (8) we obtain

$$\frac{a^3(s)}{a_0^3} \simeq \frac{a^3(s=0)}{a_0^3} + s(x,y,z)\frac{\rho_v^s(T)}{\rho_w M_0}\rho\frac{N_0}{N(x,y,z)}, \tag{9}$$

where the subscript *m* is omitted adjacent to *a* for simplicity and $N_0 = N\,(0,\,0,\,z_s)$.

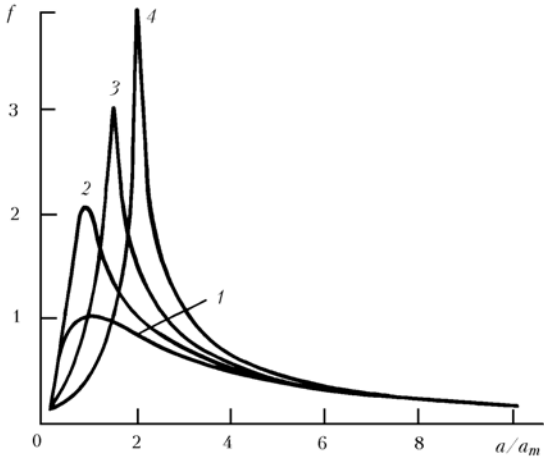

FIG. 5. Transformation of the size distribution function of aerosol particles due to their condensation growth for the degree of supersaturation s = 0 *(1),* 0.05 *(2);* 0.15 *(3), and* 0.25 *(4).*

From this it follows that

$$M_0 \simeq \frac{s(x,y,z)\rho\rho_v^s(T)}{\rho_w[a^3(s)/a_0^3 - a^3(s=0)/a_0^3]}\frac{N_0}{N(x,y,z)},$$

or taking into account Eq. (6) and ignoring the sedimentation, we obtain

$$M_0 \simeq$$

$$\simeq \frac{s(x,y,z)\rho\rho_v^s(T)}{\rho_w[a^3(s)/a_0^3 - a^3(s=0)/a_0^3]}\frac{2V_w\sigma_y\sigma_z}{R_0^2V_0}\exp\left[\frac{y^2}{2\sigma_y^2} + \frac{(z-H_0)^2}{2\sigma_z^2}\right]. \tag{10}$$

The average degree of moistening of the particles $a(s)/an$ can be determined from lidar measurements of the scattering coefficient of the system of moist particles o_s.

Within the zone of high degree of moistening $a(s)/a_0 \gg 1$ the diffraction parameter of the particles $\rho_d = 2\pi\lambda/a(s) \gg 1$ for most of the wavelengths λ of laser sources employed in lidar systems which makes it possible to set the scattering efficiency $K_s(a(s))$ equal to unity

$$\sigma_s|_{\sigma>0} = \pi N(x,y,z)\int_0^\infty F(a)a^2K_s(a)\mathrm{d}a \simeq \pi N a_m^2(s). \tag{11}$$

Combining Eqs. (10) and (11) we obtain the relation for determining M_0

$$M_0 \simeq \frac{N_0\rho}{N(x,y,z)}\left[\sigma_p\frac{4(a_m/a_0)a_0}{3} - \frac{\rho_v^s(T)}{\rho_w}s(x,y,z)\right],$$

which can be numerically calculated for the given parameters of emission.

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