Modeling the spread of aerosol cloud accompanying liquid propellant emission into the atmosphere

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We present a physical-mathematical model for describing the evolution of a cloud of toxic liquid propellant emitted in the emergency case of crashes and/or stage separation. The model developed takes into account the polydisperse composition of droplets, instability of their motion, and the prevailing wind. This model accounts for the process of droplet heating and freezing in different atmospheric layers, the phase changes, aerodynamic splitting, and turbulent diffusion. Some results calculated for a typical liquid propellant components are presented.

The stage separation of liquid-propellant carrier rockets may result in atmospheric emissions of the propellant and its oxidizer. Similar emissions of liquid-droplet components can also take place upon crashes of rockets in the atmosphere or their fall on the Earth's surface. In the latter case, the propellant components partly self-ignite, and thus produced mixture of the combustion products and liquid-droplet components ascends up to the hoverheight and then descends under the effect of gravity. The emission of these components is accompanied by a set of complex interrelated physicochemical processes (evaporation, chemical heterogeneous reactions, diffusion, coagulation, splitting of droplets, etc.). As a result, the toxic components spread in space and, finally, deposit onto the surface.

To assess ecological consequences of such events and the damage to the target territories, it is necessary to have reliable physicochemical models of the considered processes. Note that one of the most important parameters of the model is the vertical coordinate of depressurization (flight height), because all the basic environmental characteristics (density, temperature, pressure, chemical composition, direction and strength of the dominant wind) depend on this parameter. The altitude dependence of the atmospheric characteristics is studied in a sufficient detail and can be introduced into the model under development from the corresponding reference books. When constructing the physicochemical model of a spread of liquid-droplet toxic components, it is necessary to understand the following processes in detail:

1. The formation of the primary droplet cloud at the time of depressurization of fuel tanks.

2. Processes of equilibrium and nonequilibrium droplet evaporation at a sharp change of the ambient conditions due to depressurization.

3. Diffusion and spread of the components under the action of the dominant wind.

4. Chemical reactions of the propellant components with the atmospheric components.

5. Processes of gravitational sedimentation of droplets with the allowance for evaporation and splitting upon interaction with gas, as well as the processes of gravitational coagulation and splitting upon collision of different-size droplets.

The behavior of the propellant droplets (mostly, unsymmetrical dimethylhydrazine (UDMH)) falling from high altitudes was considered elsewhere.[1–4] The rate of gravitational sedimentation was calculated, as a rule, using the dependence for the stationary (established) droplet fall velocity obtained through introduction of the correction factor into the Stokes law. Analysis of the conditions of droplet sedimentation with allowance for the parameters of the standard atmosphere and the Reynolds and Weber numbers has shown that the use of the stationary dependence for the droplet fall velocity is acceptable only under conditions near the ground (the relaxation time not exceeding few seconds). At high altitudes of the fuel tank depressurization (above 50–100 km) under conditions of low air density, droplets acquire the stationary velocity upon falling a significantly long path (up to tens kilometers).

The model of evolution of a cloud of toxic monodisperse liquid-droplet components was considered in Refs. 5–7 with the allowance made for the dynamics of the droplet velocity and the refined dependence for the aerodynamic drag factor under non-Stokes conditions. Among a few experimental papers, to be noted there is Ref. 8, in which airborne studies of the aerosol wake formed due to emission of oxidizer (nitrogen tetroxide) from a falling expendable stage of a carrier rocket have been carried out.

Here we present a physical-mathematical model and some calculated results on the evolution of a cloud of typical liquid-propellant components, such

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as UDMH (heptyl), nitrogen tetroxide, and kerosene upon depressurization of the carrier rockets. This model accounts for the acceleration of polydisperse ensemble of droplets during gravitational sedimentation, droplet heating and cooling in different atmospheric layers, as well as evaporation and turbulent diffusion with the allowance for dominating wind. For obtaining an experimental confirmation of the adequacy of the model, some calculations have been performed for rain droplets.

The most difficult aspects calling for additional studies are the processes of formation of the primary droplet cloud upon fuel tank depressurization and the parameters of dynamic interaction of the droplets in the process of gravitational coagulation. Depressurization involves a significant pressure drop that causes emission of the liquid accompanied by its fragmentation and the loss of their stability yielding the formation of fine droplets. These processes result in the formation of the initial cloud with some particle size distribution function $g(r)$ (r is the droplet radius).

Since these processes are hard to describe deterministically and because they depend on a lot of random factors, in particular, on the dynamics and geometry of fuel tank depressurization, it is worth representing $g(r)$ as a lognormal distribution, following the Kolmogorov's idea.[9] In this paper, the particle size spectrum in the initial cloud was described by the Rosin–Rammler equation[10]:

$$g(r) = (n/r_*^n)\, r^{n-1}\exp[-(r/r_*)^n], \qquad (1)$$

where n, r_* are the distribution parameters.

As was shown in Ref. 11, Eq. (1) is well approximated by the lognormal distribution in a wide range of n and r_*. The function $g(r)$ is normalized to unity, that is, $\int_0^\infty g(r)\mathrm{d}r = 1$. The modal radius (corresponding to the maximum of the distribution function) is $r_\mathrm{m} = r_*/\sqrt{2}$. In this case, $g(r)\mathrm{d}r$ is the mass (or volume) fraction of droplets, whose size falls within the range $(r, r + \mathrm{d}r)$. The values of the parameters $n = 2$, $r_* = 2$ mm are chosen in accordance with the analysis of experimental data on the size of rain droplets.[12–14]

In the calculations, the distribution (1) was approximated by six fractions (Fig. 1); the corresponding sizes r_i and mass fractions $z_i = c_{\mathrm{m},i}/c_\mathrm{m}$ are summarized in Table 1 ($c_{\mathrm{m},i}$ is the mass concentration of particles of the ith fraction; c_m is the mass concentration of droplets in a cloud).

The following scenario of droplet cloud formation and evolution was considered. Upon depressurization of fuel tanks of a carrier rocket (as a result of a crash or stage separation) and splitting of the emitted liquid (of mass M) into fragments at some altitude H from the ground, a cloud of the volume V, containing the uniformly mixed droplets with the size distribution described by Eq. (1), is formed. The minimum volume of the droplet cloud V_min can be estimated by the following equation (for the case of close-packed spherical particles)

$$V_\mathrm{min} = 6M/(\pi\rho_\mathrm{p}), \qquad (2)$$

where ρ_p is the liquid density.

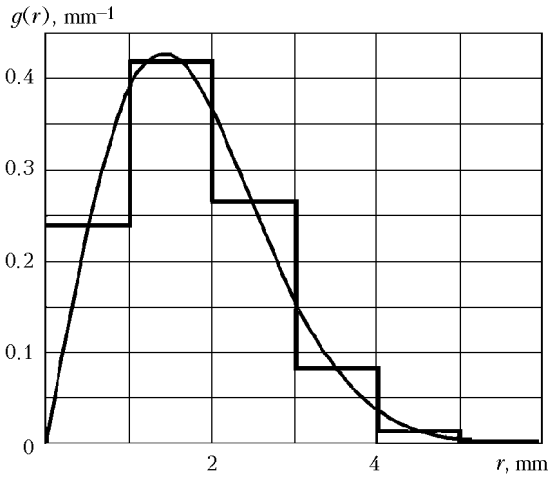

Fig. 1. Rosin–Rammler distribution.

Table 1. Initial droplet size distribution

Fraction	1	2	3	4	5	6
r_i, mm	0.5	1.5	2.5	3.5	4.5	5.5
z_i	0.231	0.422	0.258	0.078	0.010	0.001

The actual volume is larger due to droplet spreading and is determined by the equation

$$V = a^3 V_\mathrm{min}, \qquad (3)$$

where a is the coefficient accounting for the droplet spreading ($a = 1$ corresponds to the close packing; $a = 2$ is for the case that the separation between droplets is equal to their diameter; $a = 3$ is for the case that the separation between droplets is equal to the doubled diameter, and so on). Taking $a = 10$ we can estimate by Eqs. (2) and (3) the initial volume V_0 or the radius R_0 of the droplet cloud (assuming its sphericity). Thus, for example, for UDMH droplets ($\rho_\mathrm{p} = 790$ kg/m^3) at $M = 100$ kg the radius of the primary cloud is $R_0 = 3.8$ m. It should be noted that for correct choice of the coefficient a, we need *a priori* information about the process of formation of the primary droplet cloud, which can be obtained by direct observation or from solution of the corresponding inverse problem (by analogy with Refs. 3 and 4).

However, taking into account that $R_0 \ll H$, the value of R_0 should not have a strong effect on the cloud evolution due to turbulent diffusion in the atmosphere.

The equation of motion for droplets of the ith fraction can be written in the form

$$m_i \frac{\mathrm{d}\mathbf{v}_{\mathrm{p},i}}{\mathrm{d}t} = \sum_j \mathbf{F}_j, \qquad (4)$$

where $\mathbf{v}_{\mathrm{p},i}$ is the velocity vector of droplets of the ith fraction; $m_i = (4\pi r_i^3/3)\rho_\mathrm{p}$ is the mass of a droplet

of the ith fraction; $\mathbf{F}_j$ are the forces acting on the droplet.

Neglecting small Archimedes force, the equations of motion in the projections onto the axes of the Cartesian coordinate system x, y, z (the axis z is directed along the vector of the acceleration due to gravity $\mathbf{g}$) have the form[15]:

$$\frac{du_{p,i}}{dt} = \frac{3\rho}{8\rho_p r_i} C_D |\mathbf{v} - \mathbf{v}_{p,i}| (u - u_{p,i}),$$
$$\frac{dv_{p,i}}{dt} = \frac{3\rho}{8\rho_p r_i} C_D |\mathbf{v} - \mathbf{v}_{p,i}| (v - v_{p,i}), \quad (5)$$
$$\frac{dw_{p,i}}{dt} = \frac{3\rho}{8\rho_p r_i} C_D |\mathbf{v} - \mathbf{v}_{p,i}| (w - w_{p,i}) + m_i g.$$

Here

$$|\mathbf{v} - \mathbf{v}_{p,i}| = \sqrt{(u - u_{p,i})^2 + (v - v_{p,i})^2 + (w - w_{p,i})^2}$$

is the absolute value of the difference vector; u, v, and w are the components of the wind vector $\mathbf{v}$; $v_{p,i}$, $u_{p,i}$, and $w_{p,i}$ are the components of the velocity vector for droplets of the ith fraction; ρ is the air density; C_D is the aerodynamic drag factor.

For the aerodynamic drag factor, Klyachko equation[16] was used:

$$C_D = \frac{24}{\mathrm{Re}} + \frac{4}{\sqrt[3]{\mathrm{Re}}}, \quad (6)$$

where

$$\mathrm{Re} = \frac{2\rho |\mathbf{v} - \mathbf{v}_p| r}{\mu}$$

is the Reynolds number of the relative motion; μ is the coefficient of the air dynamic viscosity.

The dependence (6) is valid in the range of Re values from 1 to 700 (intermediate mode of flow), while for the range of Re from 700 to $3 \cdot 10^5$ (self-similar mode of flow) we used the value $C_D = 0.44$ (Refs. 10 and 15).

The rate of gravitational sedimentation of droplets w_{p0} was determined from solution of Eq. (3), which reduces to the transcendent equation at $d\mathbf{v}_{p,i}/dt = 0$.

Table 2 summarizes the calculated results on the rate of stationary deposition of water droplets and the corresponding experimental data on the velocity of raindrops,[12–14] along with the calculated Reynolds and Weber numbers

$$\mathrm{We} = (2\rho |\mathbf{v} - \mathbf{v}_p|^2 r)/\sigma,$$

where σ is the liquid surface tension coefficient.

The calculated results are in a good agreement with the experimental data on the droplet velocity, which is indicative of the adequacy of the dependences used in the model.

To take into account aerodynamic destruction, it was assumed that the droplet splits into two spherical droplets of equal mass as the Weber number achieves the critical value $\mathrm{We} = \mathrm{We}_* = 17$.

The effect of the vertical coordinate on the physical properties of air was taken into account using the parameters of the standard atmosphere,[17] and the dynamic viscosity coefficient was calculated by the Sutherland equation

$$\mu = \frac{0.68 \cdot 10^{-2}}{T + 122} \left(\frac{T}{273} \right)^{3/2},$$

where T is the absolute temperature of the air; $[\mu] = \mathrm{Pa \cdot s}$. The error in calculation of μ by this equation does not exceed 0.5% in the temperature range from 180 to 1400 K (Ref. 18).

Since the heat exchange occurs between the moving droplet and the medium in different atmospheric layers, the droplet temperature was calculated using the heat balance equation in the form

$$\frac{dT_{p,i}}{dt} = \frac{3}{2} \frac{\lambda}{r_i^2 \rho_p c_p} \mathrm{Nu} (T - T_{p,i}), \quad (7)$$

where c_p is the specific heat of the liquid; λ is the heat conductivity coefficient of the gas; $T_{p,i}$ is the droplet temperature in the ith fraction (averaged over the droplet volume).

The Nusselt number was calculated as[19]:

$$\mathrm{Nu} = 2.0 + 0.56 \sqrt{\mathrm{Re}}. \quad (8)$$

The equation for variation of the droplet radius due to evaporation has the form

$$\frac{dr_i}{dt} = -\frac{G_i}{4\pi\rho_p r_i^2}. \quad (9)$$

The rate of evaporation is determined by the equation

$$G_i = 4\pi r_i k^2 \frac{X}{1 - X},$$

where k is the mass transfer coefficient; X is the mole fraction of vapor of the droplet substance near the droplet surface.

By expressing X through the partial vapor pressure p_0, Eq. (9) can be presented in the form

Table 2. Parameters of stationary droplet deposition

r, mm	0.02	0.05	0.1	0.2	0.5	1.0	1.5	2.0	2.5	3.0
w, m/s	0.046	0.24	0.69	1.59	3.88	7.08	8.60	9.93	11.10	12.16
Re	0.12	1.63	9.26	42.4	258	943	1719	2645	3696	4859
We	$1.4 \cdot 10^{-6}$	10^{-4}	0.002	0.017	0.25	1.6	3.7	6.5	10.2	14.7
w, m/s (exp.)	0.05	0.27	0.72	1.62	4.03	6.49	8.06	8.83	9.09	9.18

$$\frac{dr_i}{dt} = -\frac{k}{\rho_p}\frac{p_0}{(p - p_0)}, \quad (10)$$

where p is the ambient pressure.

The mass transfer coefficient is calculated by the dependence[20]:

$$k = \frac{\rho_f D_f}{2r}\left[2 + 0.6\left(\frac{2r|\mathbf{v} - \mathbf{v}_p|\rho_f}{\mu_f}\right)^{1/2}\left(\frac{\mu_f}{\rho_f D_f}\right)^{1/3}\right], \quad (11)$$

where D_f is the coefficient of binary diffusion at the film temperature $T_f = (T_p + T)/2$; ρ_f, μ_f are the density and the coefficient of dynamic viscosity of air at T_f.

One can find in the literature a lot of methods for calculation of the diffusion coefficient in binary gas systems at low pressure,[19] but the most accurate method is the Fuller–Schettler–Giddings method, because it is based on a great deal of the experimental data.[20] The empirical correlation has the form

$$D = \frac{T^{1.75}[(M_A + M_B)/(M_A M_B)]^{0.5}}{p[(\Sigma V_A)^{1/3} + (\Sigma V_B)^{1/3}]^2}, \quad (12)$$

where D is expressed in cm^2/s; p is measured in atmospheres; T is in K; M_A and M_B are the molecular masses of the components A and B (liquid droplets and air); ΣV_A, ΣV_B are the diffusion volumes of the molecules.

Molecular diffusion volumes for different substances can be found in Ref. 20. Using Eq. (12), we have obtained the equations for calculation of the diffusion coefficient of water and liquid-propellant droplets in the air:

$$D = \frac{B}{p}\left(\frac{T}{273}\right)^{1.75}, \quad (13)$$

where $B = 0.22$ (water), 0.148 (nitric acid), 1.22 (kerosene), and 0.9 (UDMH).

In calculating the droplet trajectories, this system of equations should be completed with the kinematic relations:

$$\frac{dx}{dt} = u_p, \quad \frac{dy}{dt} = v_p, \quad \frac{dz}{dt} = w_p. \quad (14)$$

Upon substitution of the wind components u, v, and w, this system can be integrated numerically using the effective implicit difference scheme proposed in Ref. 15.

The system of equations (4)–(14) describes the motion of the center of gravity for droplets of the ith fraction with the allowance for evaporation, aerodynamic splitting, and wind drift. To solve it, we should set the initial conditions (droplet size distribution, size of the primary cloud, initial droplet velocity, depressurization height, and droplet concentration), as well as the data on the atmospheric parameters and wind velocity at different altitudes.

To determine the characteristics of droplet spread in the atmosphere due to turbulent diffusion, we used the combined Eulerian–Lagrangian approach,[15,21] according to which the equation of turbulent diffusion was solved for each droplet fraction:

$$\frac{\partial c_{m,i}}{\partial t} + (u - u_{p,i})\left(\frac{\partial c_{m,i}}{\partial x}\right) + (v - v_{p,i})\left(\frac{\partial c_{m,i}}{\partial y}\right) - (w - w_{p,i})\left(\frac{\partial c_{m,i}}{\partial z}\right) = \frac{\partial}{\partial x}\left(D_{t,i}\frac{\partial c_{m,i}}{\partial x}\right) + \frac{\partial}{\partial y}\left(D_{t,i}\frac{\partial c_{m,i}}{\partial y}\right) + \frac{\partial}{\partial z}\left(D_{t,i}\frac{\partial c_{m,i}}{\partial z}\right) - \frac{c_{m,i}G_i}{m_i},$$

where $D_{t,i}$ is the turbulent diffusion coefficient for droplets of the ith fraction.

In Eq. (15) the coordinate system is connected with the center of gravity of the ith droplet fraction. The coefficient of turbulent diffusion of the droplets was determined in accordance with the empirical equation[22]:

$$D_{t,i} = D_t \exp[-(r_i/r_m)^2],$$

where the turbulent diffusion coefficient D_t and the boundary conditions in Eq. (15) are determined in accordance with the recommendations from Ref. 21. Equations (15) were solved by the numerical method for the control volume.[25] The calculated distributions of the mass concentration of droplets $c_{m,i}(x, y, z)$ and the parameters of the primary droplet cloud were used to determine the topology of cloud spread until deposition onto the ground.

The physical characteristics of the typical components of liquid propellant needed for calculation of the droplet cloud evolution (according to Refs. 23 and 24) are presented in Table 3.

Table 3. Physical characteristics of the components at 20°C

Component	T_{boil}, °C	T_{melt}, °C	σ, N/m	ρ_p, g/cm^3
Water	100	0	$72.53\cdot10^{-3}$	1
Kerosene	147	–63	$24.0\cdot10^{-3}$	0.79
Nitric acid	84	–42	$59.0\cdot10^{-3}$	1.51
Nitrogen tetroxide	21	–11	$26.2\cdot10^{-3}$	1.45
UDMH	63	–58	$28.0\cdot10^{-3}$	0.79

The temperature dependence of ρ_p and σ was taken into account using the approximate dependences from Refs. 1 and 2.

The physical-mathematical model developed was used to calculate the trajectories of a polydisperse ensemble of UDMH droplets at the depressurization height $H = 18$ km for two states of the atmosphere presented in Table 4 (Novosibirsk and Kolpashevo, July 1, 2001).

The projections of the trajectories of droplets from different fractions onto the horizontal plane for these sites are shown in Figs. 2 and 3. As can be seen, the droplet can be spread to long distances (more than 20 km) from the point of the propellant emission.

Table 4. State of the atmosphere according to data of weather stations on July 1, 2001

Height, km	Direction, deg.	$\|\mathbf{v}\|$, m/s	T, °C
	Novosibirsk		
0	145	3	8
0.78	145	3	8
1.47	150	1	3
3.01	245	7	–7
5.60	260	15	–18
7.23	260	25	–30
9.21	275	36	–42
11.93	270	21	–43
13.87	275	16	–44
16.56	250	6	–48
18.00	260	32	–45
	Kolpashevo		
0	335	4	6
0.78	335	4	6
1.47	320	5	0
3.01	260	7	–7
5.60	225	10	–23
7.23	230	15	–35
9.21	220	12	–43
11.93	230	10	–39
13.87	235	8	–42
16.56	220	6	–46
18.00	220	13	–43

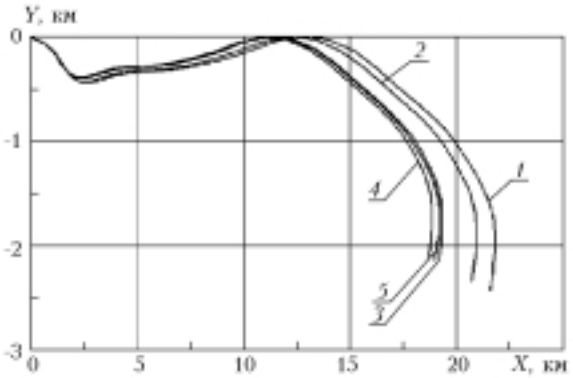

Fig. 2. Projections of the trajectories of particles from different fractions onto the horizontal plane (H = 18 km, Novosibirsk): r = 1.5 (*1*), 2.5 (*2*), 3.5 (*3*), 4.5 (*4*), and 5.5 mm (*5*).

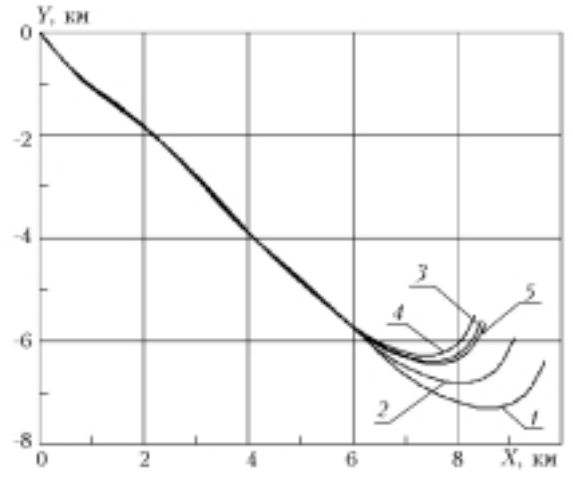

Fig. 3. Projections of trajectories of particles from different fractions onto the horizontal plane (H = 18 km, Kolpashevo): r = 1.5 (*1*), 2.5 (*2*), 3.5 (*3*), 4.5 (*4*), and 5.5 mm (*5*).

Conclusions

The paper presents the physical-mathematical model of evolution of a cloud of toxic liquid-droplet components formed upon emission of liquid propellants into the atmosphere.

This model accounts for the polydisperse composition of the formed droplets, nonstationary character of droplet motion, droplet heating and cooling processes, turbulent diffusion, and effect of the dominant wind on the topology of a local pollution.

The comparison of the results of test calculations with the data on raindrops available suggests the adequacy of the model developed.

To improve the accuracy of predicting the ecological consequences of an emission of liquid propellants into the atmosphere, it is needed to refine the size of the primary droplet cloud and to take into account gravitational coagulation upon collisions of droplets from different fractions.

Acknowledgments

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