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A MATHEMATICAL MODEL, ALGORITHM, AND PACKAGE OF PROGRAMS FOR SIMULATION AND PROMPT ESTIMATION OF THE ATMOSPHERIC DISPERSION OF RADIOACTIVE POLLUTANTS

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A mathematical model and a package of programs are presented for simulating the atmospheric turbulent diffusion of contaminating impurities from land based and other sources. Test calculations and investigations of the effect of various factors are carried out.

Introduction. The problems of nuclear safety associated with the atmospheric propagation of radioactive emissions are very important for designing new and operating the available nuclear power objects. For Belarus the problem of the secondary redistribution of radioactive contaminants in the atmosphere is particularly urgent after the Chernobyl accident. Dispersion processes in the atmosphere are associated with both the atmospheric turbulent diffusion of radioactive contaminants occurring in the air due to the secondary lift by the wind and advection of these admixtures in the atmosphere.

At the present time different approaches to the solution of the problem about the atmospheric transport of contaminants in the regional and transboundary scales are available. In principle, all models are divided into two classes: the Lagrange and the Euler models. The difference consists in the use of moving (Lagrangian) and fixed (Eulerian) coordinate systems. In the first case, the calculation of the scattering and transfer of the emission of contaminants is performed along their trajectories in the atmosphere, and in the second case it is made with respect to a fixed geoplot. By their structure and computerization, the Lagrange models are simpler than the Euler ones and have become particularly widely used in the world foreign practice. However, certain difficulties arise during the parametrization of such models. The Euler models are more complex by their very nature and require a certain background of experience for their realization. In this case, the difficulties are associated mainly with the simulation of the advection term in the diffusion equation.

Along with the typical Eulerian and Lagrangian models, there are hybrid models. Information about the mathematical simulation in the transboundary scale can be obtained from [1-11]. As applied to the problem of transboundary transfer, the models should satisfy a number of specific requirements:

• For each portion of the admixture (impurity) its association with a certain source should be fixed in order to estimate the effect of separate sources of contamination.

• The possibility of "age" estimation of each portion of the contaminating emission should exist in order to correctly account for the processes of radioactive and chemical transformation.

• The data of model calculations must be measurable in full-scale experiments; this will ensure the verification of the model.

• In the general case the model should take various physicochemical effects into account and model the propagation of various impurities.

By its complexity and accuracy the model must correspond to the accuracy of the input information: wind velocity, rates of dry and moist precipitation, etc. Comparison of these conditions with the real possibilities of obtaining information shows that a one-layer assignment of the veocity of wind at the height of 850 GPa is possible for calculating the long-range transfer, since the use of more-detailed information does not give a substantial

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increase in the accuracy, because of interpolation errors [12, 13]. Experience in modeling and the theoretical estimates [14, 11] showed that the errors in the determination of the intensity of precipitations in the fundamental time period and of their on-site localization of precipitations may lead to very large errors in calculations over short and moderate time intervals (from a day to one year) than the errors in the assignment of the wind speed.

The parameters that determine the processes of the dry and moist removal of admixtures (the rate of dry and moist sedimentation, coefficient of vertical turbulent diffusion) are determined with a limited accuracy and are subjected to appreciable fluctuations. As shown in [15], this also creates model errors of from 5 to 30% irrespective of the type of the model.

• Physicochemical processes of the transformation and removal of radioactive admixtures may be nonlinear.

Summarizing the above, we note that with the uncertainties available in the errors of the initial information and parameters of the models (for any types of models) the errors of modeling may constitute on the average up to 50% over time intervals of the order of one day and 10–15% over the time intervals of one month and above. This fact imposes a natural limitation on the complexity and detailed examination of the models used.

The first stage of the investigation described in the present article involved the construction of mathematical models, algorithms, and programs, their justification, and the execution of preliminary model calculations and estimations for the point and areal sources of radioactive contamination.

Mathematical Model. In the majority of industrially developed countries the investigation of the atmospheric transfer of contaminants (chemical and radioactive) is based on the use of statistical models using the Taylor theory of turbulent diffusion [16]. This approach is conditioned by the sufficient mathematical simplicity of the formulation of the problem and by the expedient use of programs realizing the method.

For a single point source the basic equation for a homogeneous medium can be formulated as follows:

$$\chi(x, y, z, t) = \frac{Q}{(2\pi)^{3/2}\sigma_x\sigma_y\sigma_z} \exp\left(-\frac{(x-ut)^2}{2\sigma_x^2} - \frac{y^2}{2\sigma_y^2} - \frac{z^2}{2\sigma_z^2}\right). \quad (1)$$

This formula corresponds exactly to Fick's diffusion equation:

$$\frac{d\chi}{dt} = \frac{\partial}{\partial x}\left(K_x \frac{\partial \chi}{\partial x}\right) + \frac{\partial}{\partial x}\left(K_y \frac{\partial \chi}{\partial y}\right) + \frac{\partial}{\partial x}\left(K_z \frac{\partial \chi}{\partial z}\right), \quad (2)$$

whose solution for a steady-state case with the boundary conditions

$$\chi \to 0 \quad \text{at} \quad t \to 0, \quad r \to 0;$$
$$\chi \to 0 \quad \text{at} \quad t \to \infty$$

can be written in the form

$$\chi(x, y, z, t) = Q(4\pi t)^{-3/2}(K_xK_yK_z)^{-1/2} \exp\left[-\frac{1}{4t}\left(\frac{x^2}{K_x} + \frac{y^2}{K_y} + \frac{z^2}{K_z}\right)\right]. \quad (3)$$

We note that formula (1) corresponds exactly to Fick's expression (3) provided that the standard deviation or variance σ_i and the coefficient of turbulent diffusion K_i are associated by the relationship $\sigma_i = 2K_it$. For the steady-state case, on the condition that there is a continuous emission from a source raised to the height H, the expression for the instantaneous value of concentration at the point of space (x, y, z), with account for reflection from the Earth's surface has the following form:

$$\chi(x, y, z, t) = \frac{Q}{2\pi u\sigma_y\sigma_z} \exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\{\exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right] + R_g \exp\left[-\frac{(z+H)^2}{2\sigma_z^2}\right]\right\}, \quad (4)$$

where R_g takes into account a portion of the cloud reflected from the Earth's surface.

Thus, the problem of the spatial evolution of a plume described by formula (4) is reduced, apart from a purely technical computer realization, to the problem of determining the diffusion parameters σ_i, which generally are the functions of the state of the atmosphere.

Diffusion Parameters. According to the Taylor theorem, the diffusional parameters

$$\sigma_i^2 = 2\overline{u_i'^2} \int_0^{x/u} \int_0^t R_i(\xi)\, d\xi dt \tag{5}$$

can be determined with the use of the Lagrange correlational functions R_i and fluctuations or pulsations of the instantaneous velocity u_i':

$$R_i(\xi) = \frac{\overline{u_i'^2(t)}\ \overline{u_i'^2(t+\xi)}}{\overline{u_i'^2(t)}}, \tag{6}$$

or using the concept of kinematic viscosity ν and the power-law dependence on the wind speed:

$$R_i(\xi) = \frac{1}{\left(1 + \frac{\overline{u_i'^2}}{\nu}\right)^n}, \tag{7}$$

Sutton [17] obtained the power-law dependence of the diffusion parameter on distance in the form:

$$\sigma_i^2 = 0.5\, C_i\, x^{2-n}. \tag{8}$$

In the present work we used an approach, modified by Pasquill and Gifford, to the determination of the variance σ_i according to which the values of σ_y and σ_z are determined from the power-law dependence (or from Pasquill-Gifford's tables):

$$\sigma(x) = ax^b + c, \tag{9}$$

where x is the distance (m), a and c depend on the category of the stability of the atmosphere [17].

To be used in practice, the model was suplemented so that buoyancy forces during the formation of the plume could be taken into account:

$$\Delta h = 1.5 \frac{v}{u} d \left(1 + 1.8 \frac{\Delta T}{T} d\right), \tag{10}$$

where v is the outlet vertical velocity at the source; u is the horizontal wind speed; d is the diameter of the outlet device; ΔT is the temperature difference between the surrounding medium and the source; and T is the surrounding air temperature.

In this case, the effective height H entering into expression (4) is comprised of the sum of the height of the source h and the height of the plume Δh:

$$H = h + \Delta h. \tag{11}$$

Practical calculations by the model were made on the basis of the NIKAT computer program, which is realized for IBM-compatible computers and includes a set of graphics programs to postprocess the computational information.

To take into account specific meteorological conditions and diffusion parameters for large time periods, the program provides for summation over all probable annual wind velocities and directions, probabilities of different states of the atmosphere according to Pisquill or other classifications, etc. In other words, the annual average distribution of radioactive contaminants on the Earth's surface was determined with account for the above-indicated effects by the following relation:

$$\chi_{di}(x) = \sum_{C=A}^{F} \sum_{S=1}^{n} \frac{F_{CS_d} \overline{Q}_i}{2\pi U_S x \sigma_{ZC}(x)} \left\{ \exp\left[-\frac{(V_{S_i} x/\overline{U}_{S_i} - H)^2}{2\sigma_{ZC}^2(x)} \right] + \right.$$

$$\left. + R_g \exp\left[-\frac{(V_{S_i} x/\overline{U}_{S_i} + H)^2}{2\sigma_{ZC}^2(x)} \right] \right\}. \tag{12}$$

Removal Function. To take into account the depletion of the cloud during its motion, i.e., of the effects of radiational and chemical transformation, dry and moist removal, we used the following scheme and model. We introduce the distribution function for the vertical concentration of an admixture $\Psi(z, t)$ that satisfies the solution of the diffusion equation:

$$\frac{\partial \Psi}{\partial t} = K_z \frac{\partial^2 \Psi}{\partial z^2} \tag{13}$$

subject to the boundary conditions:

$$\Psi(z, t) = 0 \quad \text{when} \quad z = 0, \tag{14}$$

$$K_z \frac{\partial \Psi}{\partial z}\bigg|_{z=0} = V_d \Psi \big|_{z=0}, \tag{15}$$

where V_d is the velocity of dry deposition; it determines the rate of absorption of the admixture on the underlying surface and varies from zero to infinity. By integrating the function Ψ over the height, we obtain the fraction of contaminating substances occurring in the atmosphere at a given time instant:

$$\zeta_d = \int_0^\infty \Psi(z, t)\, dz. \tag{16}$$

Next, we integrate Eq. (13) over z and, taking into account Eq. (16), obtain

$$\frac{d}{dt} \zeta_d(t) = K_z \frac{d}{dz} \left[\int_0^z \frac{d\Psi}{dz} dz \right], \tag{17}$$

whence, with account for the boundary conditions, we obtain

$$\frac{d}{dt} \zeta_d(t) = K_z \frac{d}{dz} \left[\lim_{z \to \infty} \Psi - \lim_{z \to 0} \Psi \right] = K_z \frac{\partial \Psi}{\partial z}\bigg|_{z=0} = - V_d \Psi \bigg|_{z=0}. \tag{18}$$

Thus,

$$\frac{d}{dt} \zeta_d(t) = - V_d \Psi \big|_{z=0}. \tag{19}$$

To determine the function $\zeta(t)$ in explicit form, we used the expression [18]

$$\Psi(z, t) = \frac{1}{2\sqrt{\pi K_z t}} \left\{ \exp\left[-\frac{(z-h)^2}{4K_z t} \right] + \exp\left[-\frac{(z+h)^2}{4K_z t} \right] - \right.$$

$$- 2V_d \sqrt{\left(\frac{\pi t}{K_z}\right)} \exp\left[-\frac{(z+h)^2}{4K_z t} + \frac{(h+z+2V_d t)^2}{4K_z t}\right] \times \operatorname{erfc}\frac{(h+z+2V_d t)}{2\sqrt{K_z t}}\Bigg\}. \quad (20)$$

Integrating $\varphi(z, t)$ over the height and performing cumbersome calculations, we obtain an expression for the function of "dry" removal of the admixture from the atmosphere:

$$\zeta_d(t) = \int_0^\infty \Psi(z, t)\, dz = \operatorname{erf}\frac{h}{2\sqrt{K_z t}} + \operatorname{erfc}\frac{(h+z+2V_d t)}{2\sqrt{K_z t}} \times \exp\frac{V_d h + V_d^2 t}{K_z}. \quad (21)$$

The function of dry removal of the admixture from the atmosphere determines the character of the interaction of the admixture with the underlying surface at $z = 0$ and connects the parameters: K_z is the coefficient of vertical diffusion, h is the height of emission, V_d is the rate of dry deposition.

To describe the processes associated with the chemical and radiational transformation of emissions and their precipitation, we used models based on simple differential equations of the first order whose solution yields the functions of the removal (depletion) of the cloud:

a) with precipitation (moist):

$$\zeta_w(t) = \exp(-\Lambda J^\alpha t), \quad (22)$$

where the intensity of precipitation $J(x, y, t)$ depends on the coordinates and real time t, while the parameters Λ and α depend on the type of the admixture;

b) chemical or radiational transformation:

$$\zeta_T(t) = \exp(-\lambda_T t), \quad (23)$$

Here λ_T is the coefficient of decomposition or chemical transformation.

The overall function of removal is determined as:

$$\zeta_\Sigma(t) = \zeta_d(t)\, \zeta_w(t)\, \zeta_T(t). \quad (24)$$

The flux of admixture through the interface, orthogonal to the axis x and intersecting it at the point $x = r$, is determined from the following relation:

$$Q_c(t) = U \int_0^\infty \int_0^\infty c(t, l, z)\, dz dl = Q_s \int_0^\infty \Psi(z, t)\, dz = Q_s \zeta_\Sigma(t). \quad (25)$$

Operative Model with Imitation of the Plumes from Sources in the Variable Field of the Wind. The relations given above for the statistical model make it possible to construct a model that imitates the development in time and space of several plumes or jets formed by several independent sources in the variable field of the wind. Since the amount of admixture in a portion is determined by its age, we may apply the above arguments to the case when the vector of the wind velocity changes in direction and magnitude. Moreover, in the model described below the change in time of the parameters of the sources of contamination is envisaged.

The synoptic information about the wind and geopotential coming from a weather station is subjected to preliminary processing, following which the field of the real wind is calculated on its basis [19]. The field of the wind at the level of 850 GPa is transformed to the velocity of the main transfer by the formula:

$$U = \frac{U_{850}}{H_{inv}} \int_0^{H_{inv}} \left(\frac{z}{z_{850}}\right)^n = \frac{U_{850}}{n+1}\left(\frac{H_{inv}}{z_{850}}\right)^n. \quad (26)$$

where H_{inv} is the thickness of the agitated layer; z is the height; z_{850} is the height of the 850 GPa level; U_{850} is the wind velocity at the 850 GPa level.

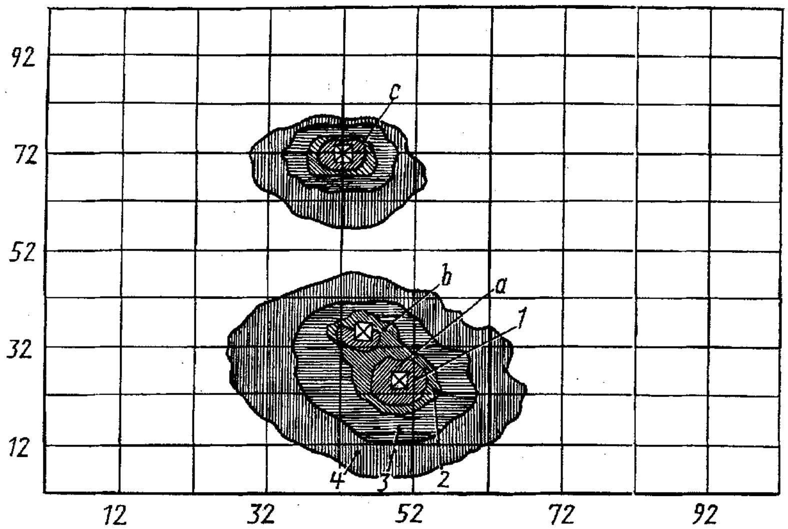

Fig. 1. Annual average distribution of the ground-level concentrations of radioactive contaminants from several areal sources (a, b, c) with contamination density of 10–500 Ci/km^2: 1) $0.2\cdot10^{-7}$ Ci/km^2; 2) $0.95\cdot10^{-8}$; 3) $0.45\cdot10^{-8}$; 4) $0.3\cdot10^{-8}$. Parameters of calculation: power 1 Ci/g; category according to Pasquill A; time of exposure 1 h; dry deposition $8\cdot10^{-3}$ m/sec; wet deposition 0.2 mm^{-1}; roughness factor 0.8 m. The values along the axes of the grid are given in km.

According to the estimates of [20], for a neutral or weakly stable stratification in the case considered n = 0.14; this makes it possible to transform formula (26) for the transfer velocity to the form:

$$U \simeq 0.9U_{850}\,. \tag{27}$$

For calculations we used the climatological data for the Gomel and Mogilev regions of Belarus [20]. Since, as follows from the analysis of the speed and rose of winds in these regions, the appearance of dust storms is possible, we also considered such versions.

The modeling of the formation and transfer of a jet or a cloud as a whole consists in the following: suppose that portions of an admixture have the shape of squares or circles (both versions are envisaged in the model) in accordance with the dimensions and form of an elementary emitter of 500 × 500 m or of radius R = 500 m. Each emitter each hour forms a portion with the mass of the admixture equal to an hour emission. This portion undergoes agitation and expansion in the field of the real wind regardless of other portions. Let us denote the coordinates of the center of the portion at the time instant t_n by x_n, y_n and the linear dimensions along the Cartesian axes X_n and Y_n by x and y. The new position of the center of the portion in time Δt can be determined from the following relations:

$$X_{n+1} = X_n + \overline{U}_n\Delta t\,; \qquad Y_{n+1} = Y_n + \overline{V}_n\Delta t\,, \tag{28}$$

where $\overline{U}_n$ and $\overline{V}_n$ are the mean weighted components of the velocity vector in the directions X and Y, respectively.

When constructing the jet, we assumed that in the first approximation diffusion in the horizontal direction is proportional to the length of the jet and is determined by the expression

$$\sigma_y \simeq 0.1R\,,$$

where R is the length of the jet axis. Next, if the Gaussian distribution is approximated by $\pm l$, uniform over the finite interval, then we may assume that $|l| = 2\sigma_y$, where $|l|$ is the half-width of the jet.

Verification of the Models, Parametric Investigations, and Analysis of the Results. As was expected, at the first stage of studies we carried out parametric investigations of the effect of various factors: speed and direction

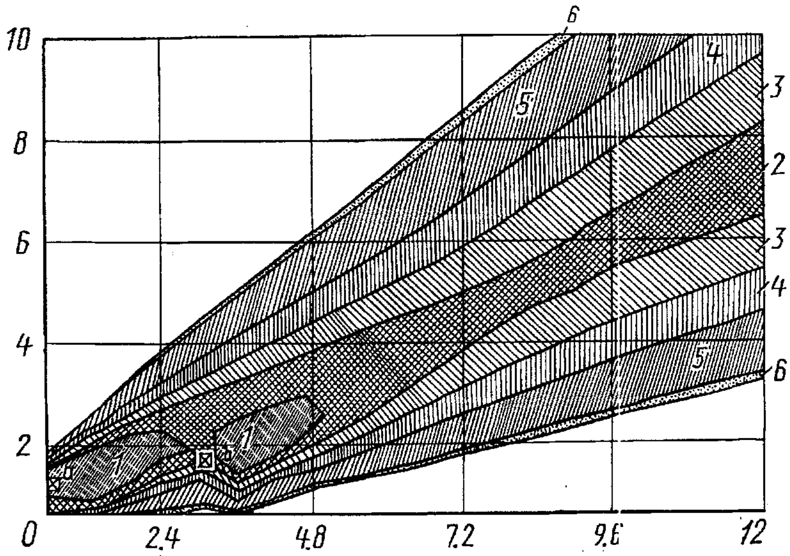

Fig. 2. Calculation of the precipitation (ground-level concentration) of contaminants from two independent sources in the variable field of the wind with account for the "dry" and "wet" removal of admixtures and for processes of radiochemical transformation of the admixture: 1) $0.1 \cdot 10^{-6}$ Ci/km^2; 2) $0.7 \cdot 10^{-7}$; 3) $0.5 \cdot 10^{-7}$; 4) $0.3 \cdot 10^{-7}$; 5) $0.1 \cdot 10^{-7}$; 6) $0.8 \cdot 10^{-8}$ Ci/km^2. Parameters of calculation: time step 0.67 h; time of exposure 2.2 h; category according to Pasquill A; dry deposition $8 \cdot 10^{-3}$ m/sec; wet deposition 0.2 mm^{-1}; height of sources: a) b) 1 m; power of source: a) 6.7 Ci/year; b) 3. Values along the axes of the grid are given in km.

of the wind, state of the atmosphere, intensity of the sources of contamination, etc. Moreover, we carried out comparisons of the values calculated by the model with the data of other authors (we solved model test problems to verify and justify the validity of the programs). For example, we used the trial function method to verify the programs and algorithms, and also to filter out syntactic errors in the codes of the programs.

Effect of Diffusion Parameters and Climatic Conditions. To determine the effect of diffusion parameters (categories of the stability of the atmosphere), we calculated the versions with different, according to Pasquill-Gifford, condtions for the annual average distribution of radioactive contaminations. A version of calculation for the concentrations of contaminants that correspond to the annual average precipitation from several areal ground-level sources (emitters) is shown in Fig. 1. We note that the calculations carried out correspond to different categories of the stability of the atmosphere. In view of the absence of reliable information about the annual average probabilities of the realization of this or another category of stability according to Pasquill-Gifford, in the present work we carried out parametric investigations of the effect of different categories.

Effect of the Parameters of Dry and Moist Precipitation. Since at the present time there are no specific real data on the time distribution of the rates of dry and moist removal of radionuclides ^{90}Sr and ^{137}Cs as well as ^{238}Pu from the atmosphere, in the present work we carried out estimations of the effect of these parameters in the range of their variation that corresponds to the change of V_d within 0.3–1 cm/sec for cesium and strontium and of V_d from 1 to 5 cm/sec for plutonium. Figure 2 shows the isolines of the concentration of radioactive contaminants with account for dry and moist removal of admixtures from a cloud. The most intense sedimentation corresponds to the version of a wet (with rain or fog) precipitation. It should be noted that such versions of the state of the atmosphere are very typical for the territory of Belarus.

The results calculated by the NIKAT program for the trajectory of the admixture cloud from two ground-level continuous sources are presented in Fig. 3. The calculation was performed for 2.2 hours with a four-minute interval. The model envisages the change in the velocity vector on each time interval, as well as the effect of dry and wet depletion of the cloud. In this case the maximum values of the lines for the level of the ground concentration amounted to about 10^{-6} Ci/km^2.

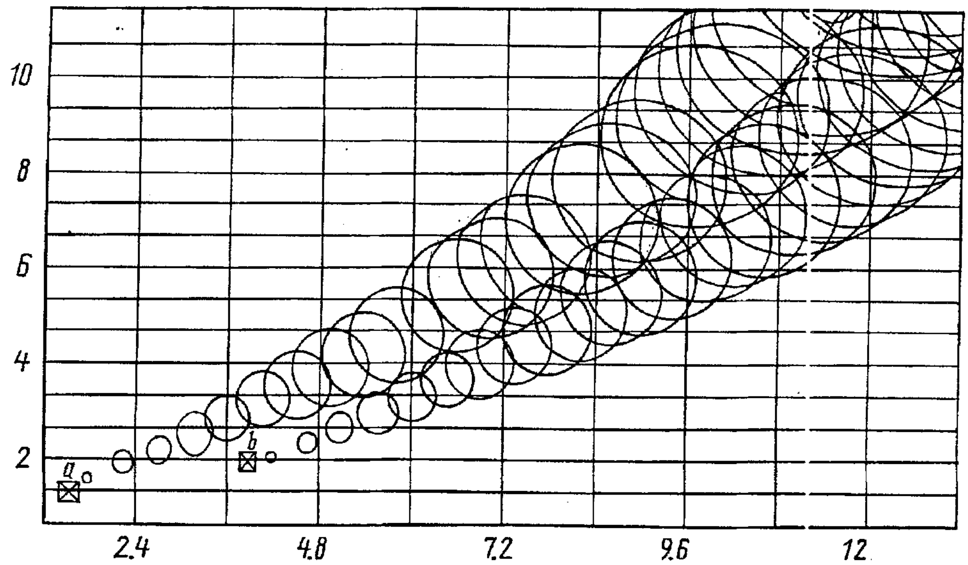

Fig. 3. Calculated trajectories of the motion of a contamination cloud. Operative model. Time of calculation 2.2 h; interval 0.07 h. Symbols are same as in Fig. 2.

Conclusion. The presented mathematical models and algorithms of the dispersion of radionuclides use the Lagrange trajectory model to describe the propagation of radionuclides in the atmosphere and permit one to carry out investigations and prediction of the effect of different factors: velocity and direction of wind, climatic conditions, intensity of wind rise, etc.

The atmospheric dispersion of radioactive contaminants is a complex physical process that depends on many factors: diffusion parameters, velocity and direction of wind, rate of dry and wet precipitation, seasonal intensity of the emitter of contamination, etc.

Preliminary calculations and investigations as well as analysis of the available data (for example, for the Karachaev accident) showed that secondary wind transfer should be taken into account when composing regulations and recommendations as well as developing a strategy for the long-term storage and handling of radioactive wastes and also predicting the state of affected objects and grounds on the territory of Belarus.

The analysis of the results of calculated and climatological data for the Gomel and Mogilev regions of Belarus as well as the study of the consequencies of the Kushtym accident proved the existence of a real danger (in the case of an unfavorable combination of climatic and atmospheric conditions) of an intense wind transfer of radioactive contaminants over the territory of Belarus and neighboring countries.

The proposed models and programs are intended for the investigation of complex atmospheric phenomena of the transfer of admixtures; in future they may furnish the basis for the investigation of synergistic effects of the simultaneous influence of chemical, radiational, and other conversions. A further refinement of the programs will make it possible to calculate human tolerance doses for population due to the effect of various factors, as well as overall contamination fluxes through the boundaries of the regions.

The NIKAT package has a modular structure, which permits one to easily readjust and adapt the programs for modeling different transport effects of contaminants in different media from different combinations of several simultaneously acting sources. Separate blocks of programs can be used for the further development of new modules and improvement of the available ones.

NOTATION

H, height of the source of emission; K_x, K_y, K_z, coefficients of diffusion; Q, power of the source; u, wind velocity; t, time; σ_x, σ_y, σ_z, diffusion parameters (standard deviations); χ_{di}, concentration of radionuclide i on the Earth's surface as a function of the distance x from the source in the sector d; n, number of the intervals of the wind velocities; $\overline{Q}_i$, annual average emission of the i-th radionuclide; $\overline{U}_S$, the mean value of wind velocity in the interval S; V_{S_i}, rate of sedimentation for the i-th radionuclide; x, y, z, Cartesian coordinates (along the wind, across the wind, in the vertical direction); R_g, correcting coefficient to account for the effect of the reflection of plume from the Earth; R_i, Lagrange autocorrelation function of velocity; $\Psi(z, t)$, distribution function of the vertical concentration of admixture; $\zeta_\Sigma(t)$, $\zeta_d(t)$, $\zeta_w(t)$, $\zeta_T(t)$, functions of the removal (depletion) of the cloud (Σ, overall; d, dry; w, wet; T, chemical and radioactive transformation); α, constant of washing-out or decomposition.

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