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Original Article

A new approach for modeling pulse height spectra of gamma-ray detectors from passing radioactive cloud in a case of NPP accident

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ABSTRACT

A comprehensive approach for modeling the pulse height spectra of gamma-ray detectors from passing radioactive cloud in a case of accident at NPP has been developed. It involves modeling the transport of radionuclides in the atmosphere using Lagrangian stochastic model, WRF meteorological processor with an ARW core and GFS data to obtain spatial distribution of radionuclides in the air at a given moment of time. Applying representation of the cloud as superposition of elementary sources of gamma radiation the pulse height spectra are calculated based on data on flux density from point isotropic sources and detector response function. The proposed approach allows us to obtain time-dependent spectra for any complex radionuclide composition of the release. The results of modeling the pulse height spectra of the scintillator detector NaI(Tl) Ø63x63 mm for a hypothetical severe accident at a NPP are presented.

1. Introduction

In accordance with the requirements [1–3], continuous radiation monitoring must be carried out on the territory in the vicinity of the NPP. The tasks of radiation monitoring include: rapid detection of significant changes in dose rates or concentrations of radionuclides in the air; provision of information and data for the assessment of radiation doses to the population, including actual and predicted values; information support in making decisions aimed at eliminating accidents [1,2]. Currently, equipment installed at existing monitoring system posts measures mainly the dose rate values in the on-line mode [4–9]. However, it does not allow us to obtain the radionuclide composition of air contamination. The use of gamma-spectrometric equipment in monitoring systems for direct measurements of the radiation from the radioactive cloud can make it possible to obtain operational data on the qualitative and quantitative nuclide composition of the cloud, type of accident at NPP that led to the release of radioactivity into the atmosphere, and its dynamics. Also, based on the analysis of the measured pulse height spectra, information can be obtained for comparison with the established limit values of nuclide concentrations in air, to adjust the operational intervention levels taking into account updated data on the accident [10,11], for taking measures to protect the population in the event of an emergency due to release of radioactivity into the atmosphere, as well as for reducing the uncertainties of the estimates of radiation doses to the population performed by calculation codes. Thus, it is an urgent task to study the capabilities of gamma-spectrometric equipment as part of the posts of radiation monitoring systems for detecting the radiation of various radionuclides contained in the radioactive cloud. This problem can be solved by mathematical modeling of the pulse height spectra of detectors measuring gamma radiation emitted by radionuclides contained in the radioactive cloud.

The issues of modeling the pulse height spectra of detectors in a case of radioactive releases into the atmosphere are considered in a number of works [12–16]. In [12], the problem of determining the reference pulse height spectra to estimate the volumetric activities of radionuclides in the air and radioactive release rate was investigated. Such reference spectra were calculated for a selected significant radionuclides in the NPP releases, using a simplified representation of the radioactive cloud based on the Gaussian dispersion model. In the study presented in [13], the pulse height spectra of the scintillation detector NaI(Tl) Ø2″x2” monitoring the radioactive releases at NPP have been modeled. Information on atmospheric transport models does not provided by authors, apparently a direct contribution from the source of the release without dilution and the dynamics of the accident was evaluated. To determine the concentrations of specific radionuclides in the air,

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including isotopes of iodine and caesium, based on the results of spectrometric analysis, the authors of the work [14] have been obtained the pulse height spectra of NaI(Tl) Ø2″x2” detector using a model of a radioactive cloud in the form of an infinite space with a uniform distribution of radionuclides. Such an approximation is correct to perform assessments outside the NPP site under unstable meteorological conditions, in some cases it is not applicable at distances up to 10 km from the source [17]. In [15], a method for modeling total absorption peak for the NaI(Tl) detector Ø2.5″ × 2.5″ mm is considered, which is one of the components of the total spectrum. The method makes it possible to calculate efficiency in terms of total absorption peak count rate using approximation of a uniform, triangular and Gaussian distribution of activity within the radioactive cloud. In [16], the total absorption peak efficiency of the NaI(Tl) detector Ø80x80 mm was calculated for a cloud in the form of a semi-infinite space, modeled by a hemisphere with uniform nuclides distribution. Thus, the methods proposed in [15,16] do not allow us to estimate the entire spectra, and use a simplified description of source of gamma radiation in air.

In this work a comprehensive approach to the determination of pulse height spectra from a passing radioactive cloud in a case of NPP accidents is proposed, based on a Lagrangian stochastic model of atmospheric dispersion, numerical weather forecast model [18] and developed model for calculating the pulse height spectra from radionuclides distributed in air. SOPRO code [19,20], developed on the basis of the previous version of the NOSTRADAMUS model [19], was used for modeling the atmospheric transport of radionuclides. Three-dimensional regional meteorological fields, obtained using the WRF meteorological processor with an ARW core [18] and constructed on the basis of the initial boundary conditions of GFS [21], were used as input data. SOPRO model in conjunction with three-dimensional meteorological data makes it possible to estimate the distribution of radionuclides in the air and their concentrations without simplified representation of the radioactive cloud, as well as to take into account the dynamics of the release and transport of radionuclides in the atmosphere. Detailed description of the physics of the atmosphere and the propagation of the cloud in the surface layer under conditions corresponding to the location of the NPP allow us to obtain time-dependent pulse height spectra at given location point near the ground surface. Also, the developed procedure for calculating the pulse height spectra is invariant to the radionuclide composition of the release, allowing us to obtain results for any combinations of radionuclides contained in the air, not limited to the specified sets of radionuclides.

To test the proposed approach the modeling of the consequences of a hypothetical severe accident at NPP has been carried out. Pulse height spectra of the scintillation detector NaI(Tl) Ø63x63mm, placed at 1 m above the ground, have been obtained for different distances from the source and time intervals after release.

2. Materials and methods

2.1. Methodology to calculate the pulse height spectra of gamma ray detectors from a radioactive cloud

The main stages of the proposed procedure to calculate the pulse height spectra of gamma ray detectors from a radioactive cloud formed as a result of an accident at NPP are shown in Fig. 1. In the first step, the radionuclide spatial distribution in the atmosphere is modelled on the basis of information on the source of the release that can be obtained from project documents or calculations using appropriate codes [22] and meteorological data in a given location. Concentration of radionuclides in the air on a 3D spatial grid are determined taking into account the time dependence.

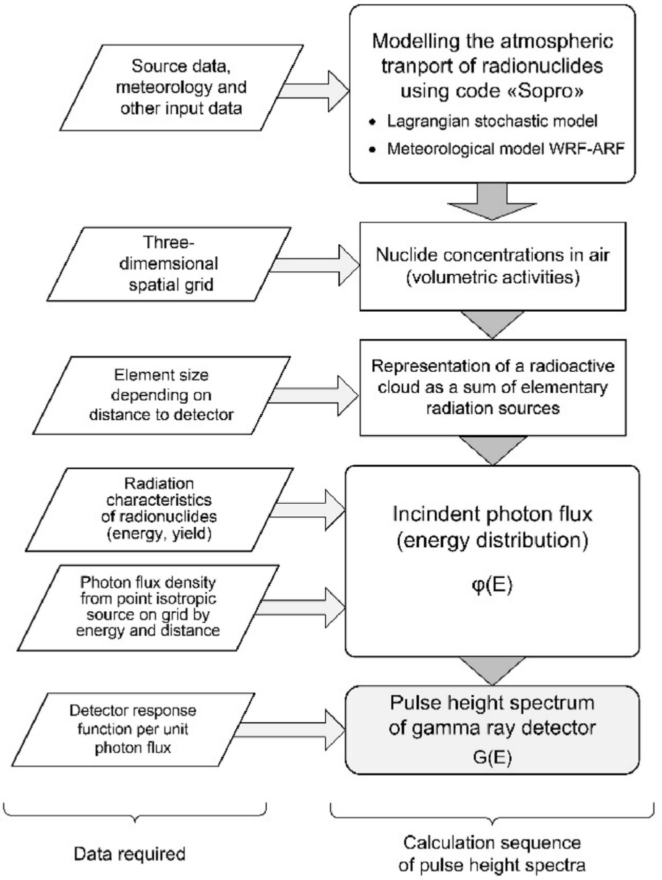

Fig. 1. Procedure to calculate pulse height spectra of gamma-ray detectors due to radiation of nuclides from passing cloud in a case of accident at NPP.

Data on the concentration and radiation characteristics of radionuclides (values of energies and yield per decay act) allow us to determine the source of gamma radiation, which will subsequently be used to irradiate detectors located at specified points near the ground surface.

Further, a volumetric source of complex shape (a cloud containing radionuclides) is divided into elementary sources, for which data on the energy distribution of the flux density for various source energies and distances to the detector locations are obtained.

Using the representation of the radioactive cloud as a superposition of elementary sources, the energy distribution of the flux density at the point of detector location is calculated.

Based on the results of calculation of the energy distribution of the flux density and response function of the detector, obtained under the conditions when monoenergetic sources of gamma radiation are placed directly at the surface of the detector, the pulse height spectrum is determined at a given location point.

2.2. 3D meteorological field model

Four-dimensional meteorological fields (three-dimensional in space and time-dependent) are used to simulate the atmospheric state changes in time and cloud transport in the computational domain. Meteorological fields serve as input data for modeling the transport of radionuclides in the atmosphere by the SOPRO code. The WRF meteorological processor with the ARW core [18] was used to calculate the meteorological fields. The initial and boundary conditions for the meteorological calculations were taken from the GFS database (USA, Global Forecast System) [21]. The GFS database resolution is 0.25 degrees, meteorological fields with a resolution of 667 m have been constructed for this work. The used grids of meteorological parameters include the following 3D values:

- wind speed projections on 3 axes;
- temperature fields;
- fields of diffusion coefficients;

as well as two-dimensional fields:

- precipitation;
- type of precipitation;
- the fact of the presence or absence of snow on the surface;
- the values of the Monin-Obukhov scale;
- height of the boundary layer;
- relative humidity at 2 m;
- air density;
- temperature at 2 m.

2.3. Model of radionuclide transport in the atmosphere

To model the transport of radionuclides in the atmosphere the code SOPRO [19,20] is used in this work, which is based on a Lagrangian stochastic transport model with the addition of a finite clouds model. This model is based on the numerical solution of the advection-diffusion equation by the Monte Carlo method.

The following assumptions are used in modeling:

- impurity transport is modeled as the transport of an ensemble of clouds;
- the trajectory of each cloud depends on the speed and direction of the wind at each point of the computational domain at the given moment of time;
- the turbulent dispersion of the impurity is modeled by applying a random displacement that depends on the state of the atmosphere;
- the change in the trajectory of one of the clouds does not affect the trajectory of the other clouds and does not depend on the results of previous calculation step.

When modeling the atmospheric transport, ingrowth and decay of nuclides in radioactive chains, impurity deposition on the underlying surface, local precipitations, advective and turbulent transport, as well as differences in the physicochemical forms of radionuclides in the release were taken into account.

In the approach used, the meteorological conditions for each cloud are determined by interpolating the values of the parameters from the model grid of the weather forecast to the point of the cloud center position.

The following formula is used for defining the concentration at an arbitrary point with coordinates x, y, z from the i-th cloud of the cloud ensemble with the center in the point with coordinates x_i, y_i, z_i:

$$
\begin{gathered}
C_i(x,y,z) = \frac{A_i}{(2\pi)^{1.5}R^2H}\exp\left(-\frac{(x-x_i)^2}{2R^2}-\frac{(y-y_i)^2}{2R^2}\right)\cdot \\
\left\{\exp\left(-\frac{(z-z_i)^2}{2H^2}\right)+\exp\left(-\frac{(z+z_i)^2}{2H^2}\right)\right\}; \\
R^2 = \int_0^t 2K_x(1-\beta)\ dt \\
H^2 = \int_0^t 2K_z(1-\beta)\ dt
\end{gathered}
\tag{1}
$$

where:

- $C_i(x,\ y,\ z)$ – concentration from the i-th cloud, Bq/m^3;
- A_i – total activity of the i-th cloud, Bq;
- x_i, y_i, z_i – coordinates of the cloud center, m;
- $x,\ y,\ z$ – coordinates of the center of an arbitrary cell, m;
- $R,\ H$ – horizontal and vertical dimensions of the cloud, m.

Three-dimensional concentration values for different time moments are obtained on a spatial grid and stored for subsequent calculations of the flux density and the pulse height spectra. The time step for calculating concentrations is 10 min. The spatial grid consists of horizontal levels located at 10, 20, 30, 40, 50, 75, 100 m and further with step of 50 m. The lateral dimensions of the cells are 100 × 100 m. The cell size of spatial grid postulated in the model is the limit for the regional atmospheric transport model determined by spatial resolution of the available data on the underlying surface with cell sizes: for terrain – 90 m, for the type of underlying surface – 500 m, for the leaf area index - 500 m, and the accuracy of the transport model, determined as a result of verification calculations.

To calculate the concentration in the elements of the 3D spatial grid, the following expressions (2) and (3) are used:

$$
\begin{gathered}
C_i^{dxdydz} = \frac{A_i}{dxdydz}\times \\
\times\left[F\left(\frac{x-(x_i-dx/2)}{R}\right)-F\left(\frac{x-(x_i+dx/2)}{R}\right)\right]\times \\
\times\left[F\left(\frac{y-(y_i-dy/2)}{R}\right)-F\left(\frac{y-(y_i+dy/2)}{R}\right)\right]\times \\
\times\left\{\left[F\left(\frac{z-(z_i-dz/2)}{H}\right)-F\left(\frac{z-(z_i+dz/2)}{H}\right)\right]+\right. \\
\left.+\left[F\left(\frac{z+(z_i-dz/2)}{H}\right)-F\left(\frac{z+(z_i+dz/2)}{H}\right)\right]\right\},
\end{gathered}
\tag{2}
$$

$$
C^{dxdydz} = \sum_i C_i^{dxdydz}
\tag{3}
$$

where:

- C_i^{dxdydz} – concentration from the i-th cloud in the reference volume V = $dx{\cdot}dy{\cdot}dz$, Bq/m^3;
- C^{dxdydz} – concentration from the entire ensemble of clouds in the reference volume V, Bq/m^3;
- A_i – total activity of the i-th cloud, Bq;
- x_i, y_i, z_i – coordinates of the center of the i-th cloud, m;
- $x,\ y,\ z$ – coordinates of the center of the reference volume, m;
- R, H – horizontal and vertical dimensions of the cloud, m;

- $F(z)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{z}e^{-z^2/2}dz$ is the error function.

In the equation (2), the concentrations in the reference volume from the i-th cloud averaged over the cell volume are recorded for the nodes of the computational grid. This calculation method allows obtaining the values of the ground-level concentration and the precipitation density that satisfy the conservation law, regardless of the ratio of the cloud dimensions and the grid cell size.

2.4. Model for calculation of the pulse height spectra from a radioactive cloud as a source of gamma radiation

Modeling the pulse height spectra of a detector includes the following main steps:

- Calculation of the energy distribution of the flux density of gamma radiation incident on the detector;
- Calculation of the pulse height spectra based on the data of the previous stage and calculated detector response function, normalized to the unit incident flux.

The representation of a radioactive cloud as a superposition of point isotropic sources is used for modeling. The energy distribution of the flux density at the detector location is calculated using the data for a point isotropic source obtained on a grid by distance and energy of the source. The energy distributions of the flux density for isotropic point sources is calculated using the Monte Carlo method [23–26] in a spherical geometry. Variance reduction techniques such as Geometric splitting / Russian roulette [26] are applied. In the intermediate points of the grid the calculations are carried out using the interpolation method. The resulting energy distribution of the flux density from radioactive cloud is calculated as a sum of contributions from all elementary sources:

$$\phi(E)=\sum_{k=1}^{Nk}\sum_{i=1}^{Nr}\sum_{m=1}^{Ni}\left[A_{k,i}\cdot\gamma_{i,m}\cdot\phi\left(r_k,E,\varepsilon_{i,m}\right)\right], \tag{4}$$

where.

- $A_{k,i}$ is the activity of radionuclide i in the discrete element k;
- $\varepsilon_{i,m}$, $\gamma_{i,m}$ – energy and yield of gamma-line m of radionuclide i;
- r_k is the distance from the discrete element k to the detector location;
- $\phi(r_k,E,\varepsilon_{i,m})$ – energy distribution of flux density at a distance r_k from a point isotropic source with the energy $\varepsilon_{i,m}$;
- Nk is the number of the discrete elements, into which the radioactive cloud was divided;
- Nr is the number of radionuclides in a given discrete element of the radioactive cloud;
- Ni is the number of gamma-lines of radionuclide i taken into account in the calculation.

This method avoids the need for time-consuming calculations to simulate the gamma radiation transport for each new geometric exposure conditions, i.e., combination of certain shape of the cloud and a given detector location. The grid by distance and energy of source of gamma radiation is chosen on the basis of ensuring an acceptable level of error while minimizing the amount of data and the cost of their preparation. The grid by distance includes values from 10 m to 1 km - 10, 25 m and further with a step of 25 m. The grid by energy of the source covers the range from 100 to 3000 keV that contains the most of the gamma-lines of known radionuclides. To divide this energy range a variable step was used: from 100 to 500 keV–10 keV, from 500 to 1000 keV–20 keV; above 1000 keV–50 keV.

The data on radiation characteristics of radionuclides were taken from the ICRP Publication 38 [27]. The width of the energy group of 1 keV is used for calculating the energy distribution of the flux density and pulse height spectra. It allows us to perform calculations for detectors with different energy resolution.

To estimate the interpolation error, we have calculated the energy distribution of the flux density in the midpoints of the grid intervals using both the interpolation and Monte Carlo methods. The calculations have been carried out for point isotropic sources containing the following gamma-emitters: Kr-87, Kr-88, I-131, I-132, I-134, I-135, Xe-138 and Cs-138. These radionuclides can be present in the air after a release from NPP and have rather sophisticated spectra of gamma radiation. The results of calculations obtained by both methods have been compared for each radionuclide. The root mean square error was used as a measure of difference. Comparison of the calculation results showed that in most cases the relative root mean square error does not exceed 15%.

After determining the energy distribution of the flux density at the detector location, the pulse height spectrum of the detector G (E) is calculated by the convolution:

$$G(E)=\int_{0}^{E_{\max}}g(E,E')\cdot\phi(E')dE', \tag{5}$$

where.

- $g(E,E')$ is the response function of the detector;
- E' is the energy of the gamma radiation incident on the detector;

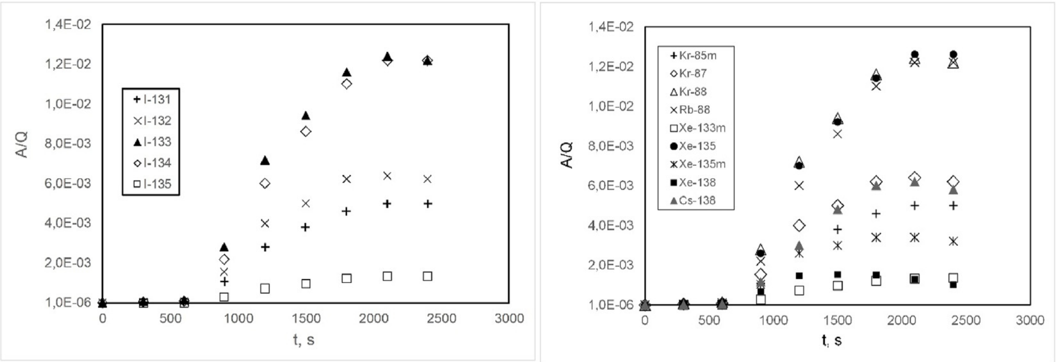

Fig. 2. Release of activity into the atmosphere as a result of accident at hypothetical NPP: (a) - iodine isotopes; (b) - other significant nuclides.

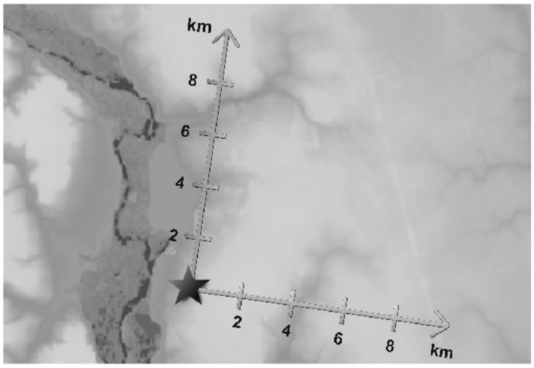

Fig. 3. Terrain in the computational domain.

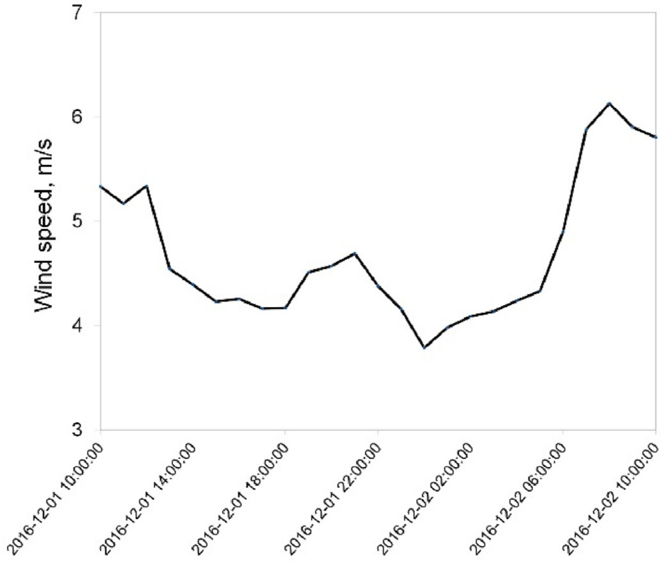

Fig. 4. Wind speed simulated by the WRF model in the location of source of release.

- $\phi(E')$ is the energy distribution of the flux density of gamma radiation incident on the detector;
- E_{max} is the boundary of the considered energy range (3000 keV).

3. Results and discussion

3.1. Results of modeling pulse height spectra from a radioactive cloud in a case of hypothetical accident at NPP

To test the developed approach to modeling the pulse height spectra and to make a primary assessment of the possibility to detect the gamma radiation of radionuclides in a radioactive cloud by gamma spectrometer, placed at 1 m above the ground, the atmospheric transport of the radionuclides in a case of severe accident at the NPP with VVER-1000 has been simulated. Hypothetical scenario with the release of radioactive substances into the atmosphere as a result of rupture of fuel element claddings and a leakage caused by a containment failure was considered. The nuclide composition of the release included isotopes of krypton, rubidium, rhodium, cesium, barium, iodine, tellurium, and xenon. In Fig. 2 the dynamics of the release of activity from the source for the considered scenario of a severe accident at NPP is shown. The values on the graphs represent the activities of the most significant radionuclides for successive 10-minute periods from the beginning of the release and are normalized to the total activity of the release $Q = 5 \cdot 10^{15}$ Bq. The activity is assumed to be released into the atmosphere uniformly within each interval.

The height of the release of activity from a hypothetical source was 40 m. A region with moderate topographical conditions was chosen as the location of the hypothetical NPP (see Fig. 3, black color - 78 m above sea level, white color - 195 m above sea level, source location is marked with star symbol, vertical axis is directed along the propagation of the radioactive cloud). The terrain along the path of radioactive cloud is flat with a height difference of less than 150 m, it includes lakes, evergreen needleleaf forests and croplands. The location of the NPP does not coincide with any of the existing NPPs. The calculated meteorological characteristics are shown in Fig. 4. The three-dimensional view of obtained radioactive clouds for different moments of time is shown in Fig. 5.

In view of the above, a simulation of radionuclide transport in the atmosphere was carried out using the SOPRO code. Using data on radionuclide concentrations in air and flux density for point sources of gamma radiation, the energy distribution of the flux density at the assumed the detector locations were calculated. After that, on the basis of expression (5), the pulse height spectra of the scintillation detector NaI(Tl) Ø63x63 mm from the gamma radiation of the passing radioactive cloud at different distances from the source of the release were obtained. The detectors were located at 1 m from the ground surface at distances up to 10 km from the source of release.

Data on the energy distribution of the flux density from point sources for distances up to 1 km have been used in the calculations, which allows us to perform estimates when the cloud passes near or directly above the detector. Thus, the detectors were placed along the cloud axis, determined by the maximum of total activity. The pulse height spectra have been obtained for time intervals up to 60 min from the start of the release with a step of 10 min. As an example, Fig. 6 shows calculated pulse height spectra of NaI(Tl) Ø63x63 mm detector for time periods 10 and 30 min and different distances from the source.

Figs. 7 and 8 show the contribution of individual radionuclides contained in the radioactive cloud to the pulse height spectra of

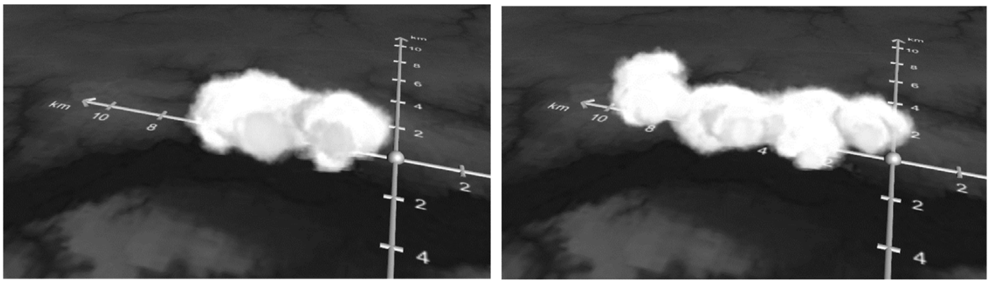

Fig. 5. 3D view of radioactive cloud at 10 (a) and 30 (b) minutes after the start of release of radionuclides into atmosphere.

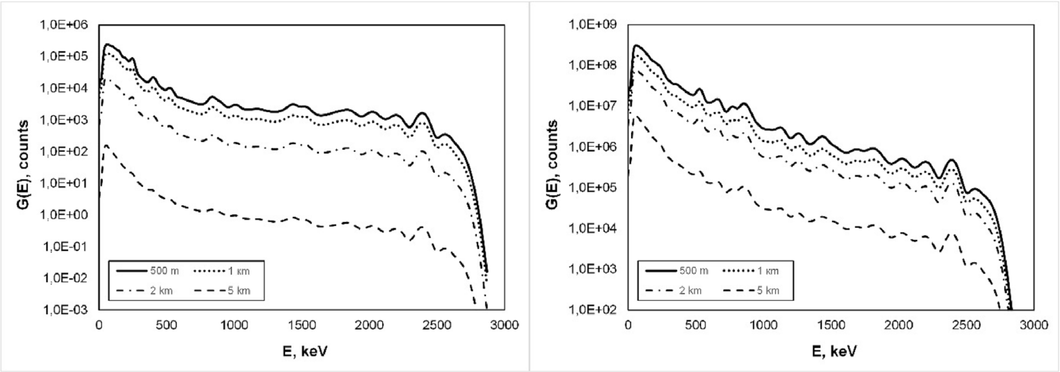

Fig. 6. Pulse height spectra of the detector NaI(Tl) Ø63x63 mm at a height of 1 m in a case of accident at NPP for 10 (a) and 30 min (b) from the start of the release for various distances from the source.

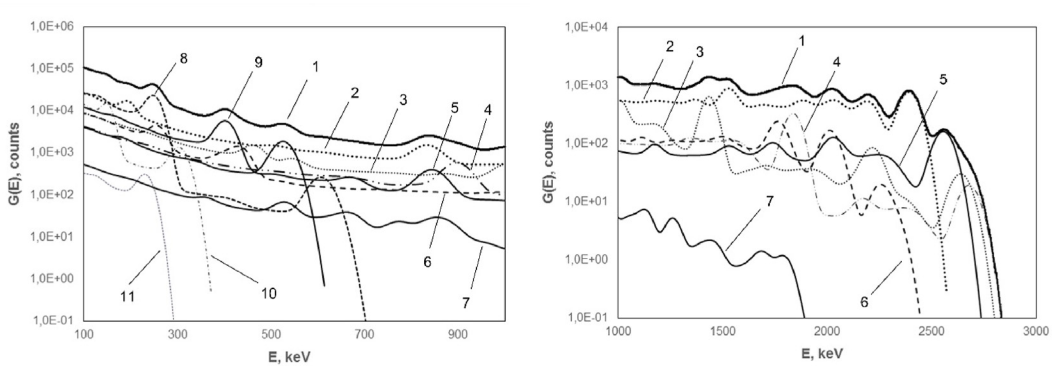

Fig. 7. Contribution of gamma radiation of different radionuclides in the radioactive cloud to the pulse height spectrum of the detector NaI(Tl) Ø63x63 mm for a period of 10 min at a distance of 1 km from the source in the energy range of 100–1000 keV (a) and 1000–3000 keV (b): 1 - total spectrum; 2 - Kr-88; 3 - Cs-138; 4 - Rb-88; 5 - Kr-87; 6 - Xe-138; 7 - Isotopes of iodine; 8 - Xe-135; 9 - Xe-135m; 10 - Kr-85m; 11 - Xe-133m.

NaI(Tl) Ø63x63 mm detector for specific distances and time periods. As can be seen, the main contribution is made by gamma radiation of the following radionuclides: Kr-85m, Kr-87, Kr-88, Rb-88, I-131, I-132, I-133, I-134, I-135, Xe-133m, Xe-135, Xe-138, Cs-138, as well as tellurium isotopes. Also one can observe total absorption peaks corresponding to the gamma-lines of Kr-87, Kr-88 and the isotopes of iodine: I-131, I-132, I-134, I-135. However, in most cases these peaks are not isolated, and this requires the application of special processing procedures [28] to resolve them. The contribution of Cs-137/Ba-137m is almost impossible to observe due to their low concentration in the air compared to nuclides listed above. In general, obtained spectra corresponded to the radionuclide composition of the radioactive cloud at the detector locations and demonstrated an adequate change in accordance with the dynamics of the radionuclide release into the atmosphere and the change of the distance from the source.

In contrast to a number of other works, the above results have been obtained without using the simplified form of radioactive

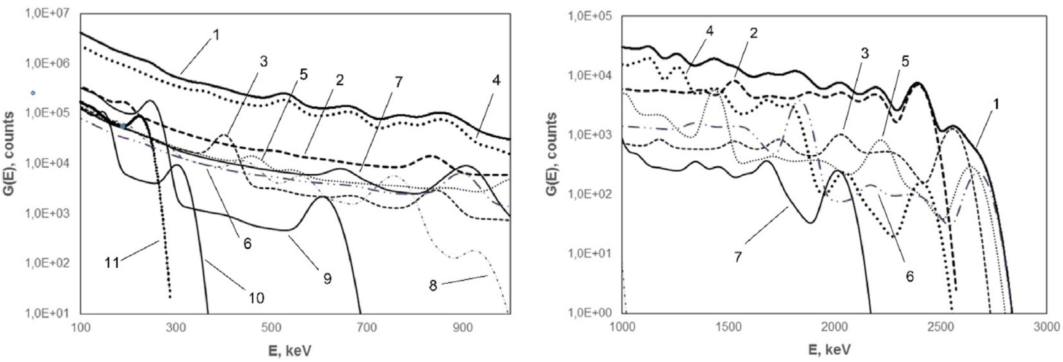

Fig. 8. Contribution of gamma radiation of different radionuclides in the radioactive cloud to the pulse height spectrum of the detector NaI(Tl) Ø63x63 mm for a period of 30 min at a distance of 5 km from the source in the energy range of 100–1000 keV (a) and 1000–3000 keV (b): 1 - total spectrum; 2 - Kr-88; 3 - Kr-87; 4 - isotopes of iodine, 5 - Cs-138; 6 - Rb-88; 7- Xe-133m; 8 - Te-133; 9 - Xe-135; 10 - Kr-85m; 11 - Te-132.

cloud and taking into account the dynamics of formation of the cloud and pulse height spectra. Work is in progress to expand data used in the algorithm to provide calculations for detectors of various types and sizes and for the cases when cloud passes aside from the detector. Study is currently being conducted to adapt the algorithm in order to evaluate contribution of radioactive fallout to the obtained spectra. It is also of interest to perform multivariate calculations using data sets with meteorological conditions in the vicinity of the source of release. This will enable us to assess the uncertainties of the results due to weather conditions.

4. Conclusion

An approach for determining the pulse height spectra of gamma-ray detectors from the radioactive cloud has been developed within this study. It allows modeling the pulse height spectra of detectors placed near the ground surface for monitoring the radioactive releases of NPP. An algorithm for calculating the spectra from the radioactive cloud as a gamma-ray source distributed in the air was implemented as an extension of the SOPRO atmospheric transport code. Calculations can be performed for an arbitrary distribution of radionuclides in the air and complex radionuclide composition of the release.

Within the framework of our research modeling of atmospheric transport of radionuclides for a postulated accident at NPP with VVER-1000 reactor has been performed. Spectra of a scintillation detector NaI(Tl) Ø63x63 mm have been obtained at various distances from the source of release and time periods up to 60 min from the start of the release. Analysis of the results showed that the main contributions to the spectra are formed by the radionuclides Kr-85m, Kr-87, Kr-88, Rb-88, Xe-133m, Xe-135, Xe-138, Cs-138 and iodine isotopes. This can be explained by their high activity in the radioactive cloud and gamma-lines with high energy. In the calculated spectra total absorption peaks from gamma radiation of a number of radionuclides can be observed, including isotopes of iodine and krypton. This allows us to conclude that it is potentially possible to determine quantitative characteristics of radionuclides containing in the radioactive cloud, even in the case of severe accident at NPP. The same calculation procedure can be applied to simulate the pulse height spectra of HPGe detectors. Primary results of our study show that no significant increase in the number of detected radionuclides is expected, although the number of registered gamma-lines increases due to the better energy resolution of HPGe detectors. This can be explained by the fact that the number of radionuclides that can be identified from spectra are determined mainly by their contribution to the total spectra, which in turn depends on relative concentration of radionuclides in air and nuclide composition of the release. Further research on the application of the proposed approach and determining the characteristics of the radioactive cloud from the obtained spectra is currently being conducted.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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