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Fuel Particles in Consequences of the Accident at the ChNPP

Ключові слова:
Chornobyl NPP,
accident,
nuclear fuel,
fuel particles,
"hot" particles,
"volatile" fraction,
acute radiation sickness.

As a result of the nuclear explosion at the fourth block of the Chornobyl NPP (ChNPP), the radioactive cloud containing an aerodispersed system with aerosols formed at the explosion appeared in air above the ChNPP territory. The accident occurred at the end of the reactor company before the assumed reloading of the active zone. Thus, the cloud composition included the products of fission and activation of uranium that were accumulated in the reactor fuel during the company. On that night, the east wind carried the radioactive cloud to the west, by leaving the aerosol fallouts on Earth's surface as a radioactive trace. The results of the own studies (1986–1990) of aerosol fallouts on the west trace formed at once after the explosion at the ChNPP are presented. On this basis, the characteristics [physico-chemical forms, radionuclidic composition, activity median aerodynamic diameters (AMADs), etc.] of the aerodispersed system created at once after the explosion of the active zone of the reactor are reconstructed. In the frame of the respiratory model given in ICRP Publication 66, the contributions caused by the inhalation of fuel particles (microscopic particles which are fragments of exploded fuel elements and have conserved mainly their radionuclidic characteristics) to the doses of irradiation of parts of respiratory organs and gastrointestinal tract (GIT) are evaluated. It is shown that the cause for a mass cough in the summers of 1986 and 1987 on territories underwent the action of emergency fallouts was the inhalation of radionuclides of ruthenium in the form of RuO_4 that was formed in "hot" particles contacting with air and then evaporated from them. The "hot" particles are compact inclusions formed by fission products. They consist mainly of atoms close to noble metals (molybdenum, ruthenium, rhodium, etc.) formed during a regular operating period in fuel tablets and released from the latter at the explosion of the active zone. The reasons for the disagreement between the clinic consequences and the ascribed values of the dose for the sufferers who were present in premises of the NPP at the emergency time and then died from acute radiation sickness in three-four weeks after the accident are explained.

Introduction

In the article, the totals of preliminary studies, whose results were published in [1–3], are presented. At first glance, it seems paradoxical that the results of measurements made three decades ago can be used for the correction of previous dosimetric evaluations and for the clarification of causes that led, in particular, to the death of some suffered persons, but this is so only at first glance. The matter is in that, by the clear causes, the characteristics of even main hazardous factors, which arose and acted at once after the explosion of the reactor, were unknown. Due to the practically complete absence of the results of corresponding measurements, the interrelations and significance of the hazardous factors were evaluated by their action on the suffered persons. It is obvious that,

in this case, somewhat was lost, and this affected the quality of dosimetric evaluations. In the first turn, unknown were the values of the radioactivity of inhalated substances and such characteristics as the physico-chemical forms of aerosols, their dispersion, and even the radionuclidic composition. Thus, the consideration "did not noticed", for example, the "transit", namely the purification of the extrathoracal and bronchial parts of respiratory organs by means of the transition of the inhalated radioactivity into gastrointestinal tract.

The retrospective reproduction of post-accident conditions, which arose at the explosion for several minutes, held for the first tens of minutes, and determined, in fact, the subsequent fate of the persons, who were present at the NPP or in its vicinity, turned out the problem far from being simple. Some very essential aspects of the event were evaluated by the results of measurements executed on the traces of the first ejection at distances of more than 1,000 km from the NPP in months and even years after the accident. By that time, the "eventual" conclusions had already been made and obtained the status of unalterable axioms by now with all following consequences. Nevertheless, some conclusions about the causes for certain clinic manifestations of the ionizing radiation action that were obtained by the results of measurements not entered the "official" annals, as well as the following consequences, seem to be of interest and can turn out the necessary supplement to and even a correction of the existing viewpoint.

It is worth to mention the existing productive experience of a similar activity: in due time, the dosimetric evaluations of radiological consequences of the atomic bombardment of the cities of Hiroshima and Nagasaki were recalculated several times for practically three decades on the basis of the gradually revealing refinements of the available results. There were no direct measurements, and they could not be made. So, the dosimetric evaluations were performed by the results of indirect measurements and the analysis of clinic manifestations of the acute radiation sickness (ARS) in the suffered persons. In this relation, the situation formed at the Chornobyl NPP at once after the explosion is similar to that in Hiroshima and Nagasaki.

Reactor before the accident

The starting prerequisite in [1–3] consisted in that the main characteristics of reactor's active zone before the accident must regularly manifest themselves in the physico-chemical and radionuclidic characteristics of emergency ejections and, by this, must determine consequences of the accident to a significant extent.

A nuclear power reactor RBMK-1000 has a thermal power of 3,200 MW. It is a heterogeneous channel thermal-neutron unit, where uranium dioxide slightly enriched by ^{235}U is used as a fuel. Graphite is a moderator, and boiling light water is a heat carrier. The accident happened at the fuel life-time end prior to the assumed recharge of the active zone.

At that time, the fuel channels of the reactor contained 1,659 fuel assemblies with 18 fuel elements in each one. To ensure a constant nominal power during the fuel lifetime, the power generation of fuel assemblies was regulated by means of their rearrangement in the active zone and by the change of "burnt" fuel assemblies by new ones. To the time of the accident, the power production of 1192 fuel assemblies was from 10.53 to 14.04 MW · day kg^{-1}, whereas 467 fuel assemblies gave from 0 to 10.52 MW · day kg^{-1}. Due to this circumstance, the data presented in Table 1 are results of the averaging of sufficiently variable values of the specific activities of rns in fuel assemblies with different power productions, which is seen on the plots in Fig. 1. Figure 1 presents the results of calculations of the ratios of specific activities for some rns in fuel assemblies with different power productions and the corresponding values of the specific activity of ^{144m}Ce. The ratio for rns ^{137}Cs and ^{134}Cs presented in Fig. 1 was used as the "cesium" ratio for the determination of the power productions of fragments of a dispersed fuel in fallouts.

The mentioned circumstances became the cause of a sort of the "macroscopic" inhomogeneity of the rns specific activity values in fuel elements, being in different fuel assemblies. Together with the "macroscopic" cause, the more essential role in consequences of the accident was played by "microscopic" inhomogeneities of the distributions of fission products (FPs) and the rns activation in a fuel of the pre-accident reactor.

Fuel elements of RBMK-1000 reactor are long metallic tubes produced of a zirconium alloy with an internal diameter of 13.6 mm containing fuel pellets. The pellets are fabricated by the pressing of uranium dioxide microgranules ~6 μm in size. The case of each fuel assembly contains 18 fuel elements. Through the fuel assemblies, water of the first contour is flowing. The heat release on the surface of fuel elements transforms water in a steam-water mixture at a pressure of ~70 atm and a temperature of ~300 °C. Then steam is separated from water and is supplied to the turbine of an electrogenerator. The temperature of the central part of pellets is as high as ~1,800 °C and decreases down to ~700 °C at their periphery.

Table 1. Ratios of the specific activities of radionuclides (rns in what follows) averaged over the active zone to the specific activity of ^{144}Ce in the fuel in 24 h after the catastrophe [4] at the mean value of the specific activity of ^{144}Ce equal to 1.74 · 1,010 Bq g^{-1}. As of the time moment of the accident, the mean beta-emission energy was $\bar{E}_\beta \approx 340$ keV

Radionuclide	Ratio, rns	Radionuclide	Ratio, rns	Radionuclide	Ratio, rns	Radionuclide	Ratio, rns
^{144}Ce	1.0	^{132}I	0.97	^{110m}Ag	0.0072	^{151}Pm	0.076
^{80}Sr	0.82	^{133}I	0.66	^{111}Ag	0.038	^{151}Sm	0.00032
^{90}Sr	0.055	^{135}I	0.083	^{112}Ag	0.0066	^{153}Sm	0.43
^{91}Sr	0.14	^{134}Cs	0.038	^{115}Cd	0.0045	^{154}Eu	0.0021
^{90}Y	0.06	^{134}Cs	0.19	^{115m}In	0.0048	^{155}Eu	0.014
^{91}Y	1.1	^{136}Cs	0.073	^{121}Sn	0.0028	^{156}Eu	0.55
^{95}Zr	1.4	^{140}Ba	0.90	^{122}Sb	0.0021	^{157}Eu	0.019
^{97}Zr	0.41	^{140}La	1.6	^{124}Sb	0.0023	^{160}Tb	0.00097
^{95}Nb	1.3	^{141}La	0.012	^{125}Sb	0.0048	^{161}Tb	0.0012
^{95m}Nb	0.0086	^{141}Ce	1.4	^{127}Sb	0.052	^{237}U	0.29
^{96}Nb	0.0027	^{143}Ce	0.72	^{128}Sb	0.0012	^{238}Np	0.15
^{97}Nb	0.45	^{142}Pr	0.52	^{129}Sb	0.003	^{239}Np	5.0
^{97m} Nb	0.41	^{143m}Pr	1.4	^{127}Te	0.059	^{243}Pu	0.0014
^{99}Mo	1.2	^{144}Pr	1.0	^{127m}Te	0.0097	^{238}Pu	0.00020
^{99m}Tc	1.2	^{144m}Pr	0.014	^{129}Te	0.033	^{239}Pu	0.00021
^{103}Ru	1.1	^{145}Pr	0.034	^{129m}Te	0.045	^{240}Pu	0.00044
^{105}Ru	0.0093	^{147}Nd	0.52	^{131}Te	0.017	^{241}Pu	0.050
^{106}Ru	0.3	^{147}Pm	0.029	^{131m}Te	0.076	^{242}Pu	0.00000059
^{105}Rh	0.41	^{148}Pm	0.076	^{132}Te	0.50	^{241}Am	0.000038
^{106}Rh	0.32	^{148m}Pm	0.069	^{130}I	0.0041	^{242}Cm	0.0086
^{109}Pd	0.052	^{149}Pm	0.45	^{131}I	0.59	^{244}Cm	0.000044

Note. In what follows, the values of activities of rns and their ratios are given by their decay to 26.04.1986.

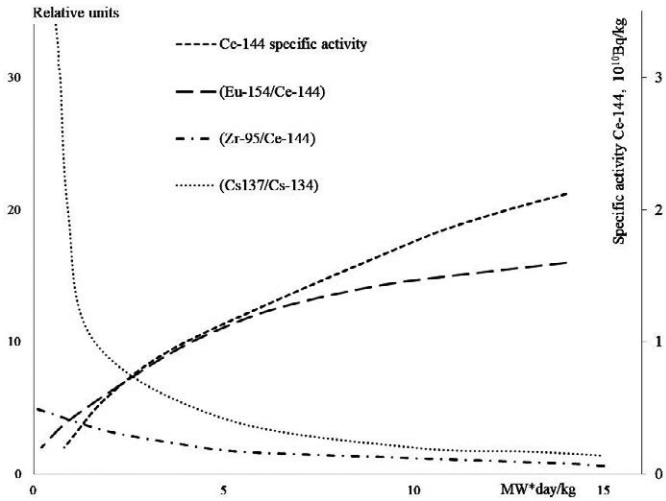


Fig. 1. Value of the specific activity of ^{144m}Ce and the ratios for some rns in the pre-accident fuel in fuel assemblies with different power productions as of 26.04.1986

The path of atoms of FPs in the substance of a fuel pellet is about 10 μm. Thus, atoms of FPs can come out during the normal operation of the reactor from uranium dioxide microgranules in intergranular cavities and can form the inclusions of FPs there, which leads to the redistribution of FPs in the pellet bulk and to the appearance of "microscopic" inhomogeneities of the distribution of FPs in a fuel.

Evaluation of the explosion power output

In the description of kinetic consequences of the accident, we mention the "Elena" plate aimed at the biological protection, which turned out on its edge in the vertical position after the reactor explosion. The plate is a disk ~$2.5 \cdot 10^6$ kg in mass and ~20 m in diameter. The minimum energy required for such event to occur should be ≈ $2.5 \cdot 10^8$ J.

Simultaneously, the numerous metallic constructions were broken, the reactor room roof was destroyed, the significant part of the active zone was ejected onto the machine room roof and on the adjacent territory, etc. If we assume that the "energy expenditures" for "Elena" are ~10^{-3} of the total explosion energy release, then the total accident energy release would be about ~$2.5 \cdot 10^{11}$ J.

We note that the nominal power of the reactor is $3.2 \cdot 10^9$ J/sec. In other words, the amount of uranium,

which would be fissionable for ~100 sec in the norm, had made it for several microseconds.

From whence, it follows that the contribution of the activity of fission products formed at the explosion had no noticeable effect on the radionuclidic characteristics of the reactor which are presented in Table 1.

Input preconditions

At once after the explosion, the ejection flare up to several hundred meters in height was formed above the reactor breakup. The radius of the spreading of fragments of the active zone, which fell on land and on the roofs of station buildings, was more than one hundred meters. A large part of debris was composed by fragments of fuel assemblies with irradiated fuel, pieces of the graphite stacking, structural elements, and settled down fragments of dispersed fuel pellets.

The dose field of photonic radiation on the debris spread territory was formed at once after the explosion and became the main cause, together with the dose from the external radiation from the ejection flare, for the subsequent death (during the spring and summer of 1986 at Hospital no. 6 in Moscow) of persons, who were present on the NPP territory outside the station premises at night after the explosion. Running ahead, we may assert with regard for the individual values of the external radiation dose that, against the background of individual values of the external radiation dose, the internal irradiation, whose doses were calculated by the results of postmortal measurements of the activity in lungs, was not able to essentially affect the fate of those persons.

As for the internal irradiation caused by the inhalation entrance of the activity, the destructions induced by the explosion resulted in that the ventilating pipe 110 m in height turned out to be directly connected at once after the explosion through certain NPP premises, where some persons were present, with the reactor breakup. The "draft" has dragged the gaseous aerosol ejection through those premises approximately for 10 min. Then the east wind "flicked" the emergency ejection flare from the Chornobyl NPP territory. Moreover, the persons in some NPP buildings, in addition to the "draft" air, breathed the air with a dispersed fuel supplied by the customary ventilation, which was operating well at the time moment of the explosion and after it.

After the accident, more than one hundred persons were delivered in Hospital no. 6 of the Health Ministry of the USSR, who were on the Chornobyl NPP territory and its premises at the night from April 26 to 27 [6].

The results of measurements gave:

hematologic estimations of individual values of the absorbed dose by the results of measurements of blood samples taken in the first days after the arrival of persons at the hospital;

results of life-time measurements of the activity of samples of urine and blood;

results of measurements of the specific activity of rns in postmortal samples of organs and tissues of the suffered persons died at the hospital in spring and summer of 1986;

values of the survival time, i. e., the number of days, for which the person has lived from the time moment of the reception of an external radiation dose and the inhalation entrance of radioactive substances at the night from April 25 to 26 till the death.

To carry out the evaluations of the internal irradiation dose, it was necessary together with the use of measurements at the hospital to extrapolate the results of studies of the emergency aerosol fallouts onto the characteristics of the aerodispersed system, which determined the entrance of radioactive substances into the persons in the premises and on the NPP territory after the explosion.

The studies of fallouts [5, 7–9] showed that the pre-accident history of a fuel and the conditions during the explosion resulted in the formation of a gaseous aerosol system in the primary ejection. This system included:

particles of a dispersed fuel (fuel particles), which are insoluble in the fluids of organism, with sizes from fractions of μm to fractions of mm and with the bulk activity of order of 1 Bq μm^{-3}. The particles conserved mainly the radionuclidic characteristics and the values of the specific activity of fuel elements, whose explosion has formed those particles;

"free" atoms and molecular compounds of the vapors of rns of "volatile" elements. A part of their atoms in fuel pellets "came out" from fuel granules into intergranular cavities of fuel pellets during the standard operation of the reactor and flied out together with a dispersed fuel at the reactor explosion (atoms of radioactive inert gases (RIGs) and mainly atoms of iodine, tellurium, and cesium);

particles of condensation ("hot" particles) with maximum size up to several μm and a bulk activity ~50 Bq μm^{-3} formed in intergranular cavities of fuel pellets during the standard operation of the reactor due to the "output" from fuel granules and the subsequent "freezing" of the atoms of fission fragments (elements with high boiling temperature such as ruthenium, rhodium, molybdenum, technetium, silver, and elements close to them) in intergranular cavities [9].

Thus, the inhalation entrances of rns of cesium, iodine, and tellurium occurred in the composition of fuel particles (insoluble form) and as separate atoms and low-molecular compounds (soluble form).

Radionuclidic ratios in fallouts

First, after the accident, more than 30 fission-produced and activation rns were registered in gamma-spectrometric measurements of the samples of air and fallouts. From them, we can separate a "nonvolatile" group including rns of zirconium, niobium, cerium, europium, and neptunium and a "volatile" one with iodine, cesium, and tellurium. The RIGs are not considered in this context, because it is assumed that they are completely evaporated at the reactor explosion. Due to the decay, ^{95}Zr, ^{95}Nb, ^{144}Ce, ^{144}Pr, ^{154}Eu, ^{155}Eu, ^{90}Sr, ^{90}Y, ^{106}Ru, ^{106}Rh (beta-emitting "nonvolatile" rns), transuranium elements, and ^{134}Cs and ^{137}Cs ("volatile" ones) remained to autumn of 1987.

It is worth to mention, as a lucky methodical finding in the studies of fallouts, the use of broadly spread handmade wooden benches at villages and small settlements as a palliative of the sedimentation samplers of aerodispersed samples [10, 11]. In the autumn of 1986 at 150 settlements that undergone the emergency fallouts from ChNPP, the measurements of the counting rates of alpha-radiation from the surface of benches were carried out by handheld alpha-radiometers.

The detailed description of the studies of benches can be found in articles and reports. In the autumn of 1987 at each of more than 50 settlements of the ChNPP zone, two fragments of benches ~15×10 cm^2 in size were taken off and composed the bank of aerosol fallouts (BAF). It was stored at the All-Union Sci. Res. Institute of Phys.-Techn. Radiomeasurements (VNIIFTRI, Mendeleevo, Moscow region) and, in essence, was the unique collection of thin-layer sedimentation samples of fallouts. All samples were measured on a PPD gamma-spectrometer, some part was studied on a scintillation beta-spectrometer and a scintillation alpha-radiometer, and some samples were used for the contact radiography with X-ray films.

The results of measurements of the samples from BAF on a PPD gamma-spectrometer and the samples of soil executed on PPD gamma-spectrometers in other organizations showed that, on the whole, the radionuclidic composition of the dispersed fuel corresponds to the averaged characteristics given in Table 1. But the following essential distinction was observed: the studies of fallouts showed that the fuel component of fallouts corresponds to the fuel of the pre-accident reactor with the burning depth approximately by 20 % less than, on the average, for the pre-accident reactor. In other words, it is related to the preferential dispersion and, hence, to the "participation" in the explosion of the content of fuel assemblies of the last charges.

Table 2 presents the radionuclidic ratios obtained by the gamma-spectrometry for the samples from BAF. It is seen that the zirconium ratios for "nonvolatile" rns are practically invariable from sample to sample, independent of the distance, and close to their fuel values from Table 1. The same is true for the ratio of rns of cesium between themselves. At the same time, the ratio of the activities of rns ^{137}Cs/^{95}Zr at large distances exceeds significantly the fuel value, decreases with the distance to the reactor, by approaching the fuel one, and fluctuates strongly from sample to sample.

Since the values of the ratio of rns ^{144}Ce/^{95}Zr are independent of the distance in the limits up to 60 km, the values of the activities of rns ^{144}Ce and ^{95}Zr are equivalent as a quantitative characteristic of the fuel component in fallouts in this zone.

Table 3 shows the data for five samples from BAF selected only near ChNPP. As distinct from the data presented in Table 2, the zirconium ratios for cesium in Table 3 do not fluctuate practically and are even less in magnitude than the fuel ones in Table 1. From whence, we may conclude that the samples represented in Table 3 contain the dispersed fuel somewhat depleted in "volatile" rns. We note that the share of evaporated intergranular inclusions in fallouts increases with the distance from the reactor, by causing an increase in the contribution of "volatile" rns revealed in the data in Table 2. Therefore, we conclude that, after the explosion, all components of the aerodispersed system were flying together. In addition, the content of radioactive cesium in the "true" fuel was approximately twice less than the rated value, by corresponding to the data

Table 2. Radionuclidic ratios for samples from BAF on various distances from ChNPP in the logarithmically normal representation. In brackets, we give β_g; lower numbers show the sampling size

Distance, km / Radionuclides	3÷17.9	18÷29.9	30÷60
^{144}Ce/^{95}Zr	0.60 (1.1) 13	0.60 (1.1) 31	0.59 (1.1) 7
^{154}Eu/^{95}Zr	$0.90 \cdot 10^{-3}$ (1.3) 12	$1.1 \cdot 10^{-3}$ (1.3) 20	—
^{137}Cs/^{95}Zr	0.051 (1.7) 13	0.10 (2.6) 31	0.46 (9.8) 9
^{137}Cs/^{134}Cs	1.88 (1.05) 13	1.85 (1.05) 28	1.76 (1.06) 7

Table 3. Typical radionuclidic ratios near ChNPP in the samples from BAF. In brackets, we give the mean square deviations (MSD) in %

Direction	W	NW	NE	S	SW
Distance, km	3	7	7	5	8
^{144}Ce /^{95}Zr	0.63 (4)	0.67 (5)	0.59 (5)	0.63 (5)	0.59 (5)
^{154}Eu/^{95}Zr	$0.94 \cdot 10^{-3}$ (15)	$0.8 \cdot 10^{-3}$ (15)	$1.1 \cdot 10^{-3}$ (15)	$0.88 \cdot 10^{-3}$ (10)	$0.71 \cdot 10^{-3}$ (10)
^{137}Cs/^{95}Zr	0.034 (5)	0.044 (4)	0.046 (5)	0.040 (4)	0.035 (5)
^{137}Cs/^{134}Cs	1.9 (4)	1.8 (6)	1.7 (9)	1.9 (5)	1.9 (4)
^{125}Sb/^{144}Ce	$4.5 \cdot 10^{-3}$ (12)	$4.9 \cdot 10^{-3}$ (20)	$2.5 \cdot 10^{-2}$ (23)	$5.2 \cdot 10^{-3}$ (20)	$4.3 \cdot 10^{-3}$ (20)

in Table 1. In other words, approximately a half of cesium was ejected from the reactor in the form of "free" atoms.

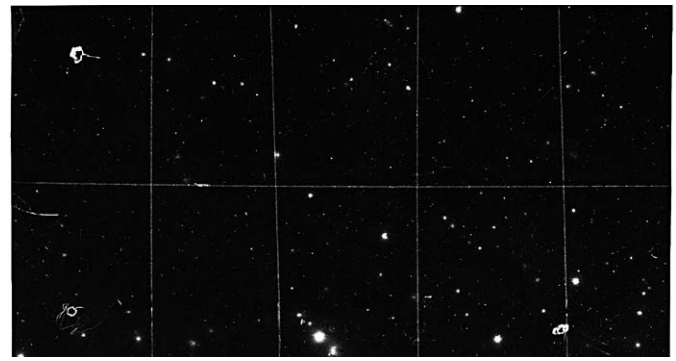
Fuel particles

In Figs. 2 and 3, we show the positive imprints of contact radiographic patterns of the beta-emission of the samples from BAF on an X-ray film that are characterized by the ratios of the activities of rns ^{137}Cs/^{95}Zr equal to 0.065 for the sample in Fig. 2a, b and to 1.07 and 5.5 for the samples in Fig. 3a, b. The radionuclidic composition of the sample in Fig. 2a, b turns out to be equal practically to the fuel one for all radionuclides and even for ^{137m}Cs. The samples in Fig. 3a, b are enriched by ^{137m}Cs to different extents. In other words, the sample in Fig. 2a, b corresponds to a practically pure fuel, whereas the samples in Fig. 3a, b are enriched with cesium from intergranular inclusions, which was concentrated for the 1.5-year stay in the open air in depressions of desks, by repeating the timber texture.

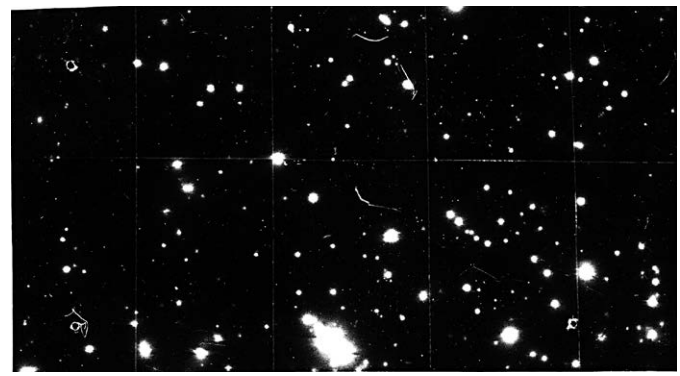
The samples from BAF, which are characterized by the zirconium ratio for ^{137}Cs close to the fuel one, were taken for the photoradiometric investigation. The scanning of their surface with a beta-counter collimated down to a diameter of 5 mm showed that practically the whole activity of beta-emitting rns of these samples is concentrated at points of their surface that gave the visible imprints on radiographic images. The distributions of the activities of alpha-emitting rns over the surface of the samples from BAF were studied, by performing the coordinate-depending photoradiometric measurements. On the imprints, which are similar to Fig. 2a, b and are "tied" to the surface of samples, we made holes ~3 mm in diameter at the irradiated places transforming the photoimprint of the sample into an "integral collimator" of its alpha-radiation.

The measurements of the counting rate of the alpha-radiation from the surface of samples from BAF covered with "collimators" and without them showed that the activity of alpha-emitting radionuclidic samples is concentrated at the flash points. Thus, the executed studies showed that the aerodispersed system forming the emergency fallouts from ChNPP contained "insoluble" particles of a dispersed fuel that were formed during the explosion, as well as the fractions formed from a substance of inclusions in this case.

The studies indicate that the value of the ratio of rns ^{137}Cs/^{134}Cs in the samples from BAF not enriched by cesium is the same in all samples, equal to the fuel one from Table 2, and does not vary over samples, despite the strong dependence on the energy production (see Fig. 1). This

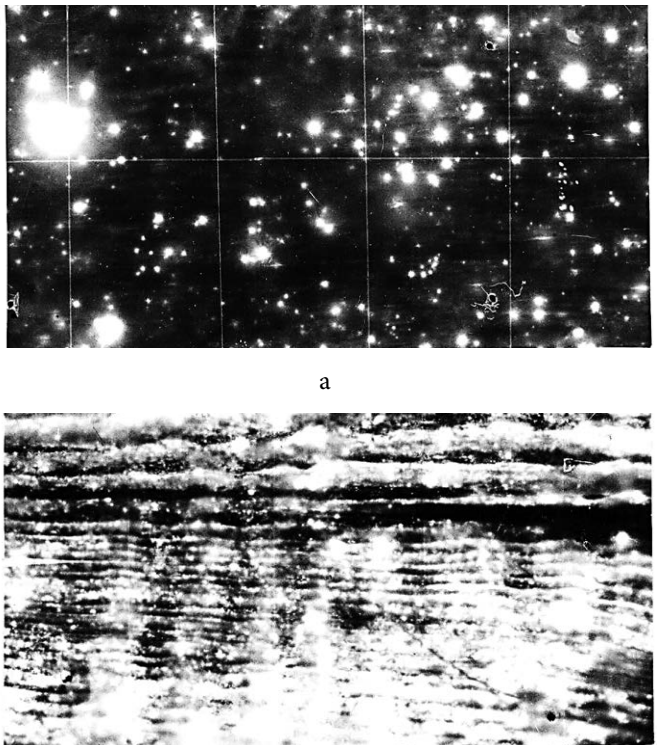


a



b

Fig. 2. Positive imprints from the radiographic images of a sample from BAF (January 1988) taken at a settlement of Chikalovichi (September 1987). Ratio ^{137}Cs/^{144}Ce of their activities is 0.065 (as of 26.04.1986). The exposure times are, respectively, 1 h (a) and 13 h (b)



a

b

Fig. 3, a, b. Positive imprints from the radiographic images of samples from BAF (January 1988) taken at the settlements of Pirki (a) and Vesnyane (b) characterized, respectively, by values of $^{137}Cs/^{144}Ce$ equal to 1.07 and 5.5 (as of 26.04.1986). The exposure time is 13 h

implies that, in the samples from BAF, the relative contribution of fuel particles formed from fuel assemblies with different energy productions coincides completely with their representation in the entire reactor as of 26.04.1986. The studies allow us to use the data of mobile measurements of the counting rate of the alpha-radiation from the surface of benches, which were carried out in the autumn of 1986, for the construction of the map of fallouts of the dispersed fuel.

For this purpose, we calculated the values of the density of fallouts of fuel particles [g km^{-2}] for 150 settlements positioned in the zone of emergency fallouts from ChNPP at distances up to 80 km from the NPP.

On the basis of the executed studies, a photoradiometric method (PRM) of measurements of the characteristics of fuel particles settled down on the surface of the samples from BAF. The method allowed us to determine, on the basis of the counting rate of the beta-radiation from the surface of the samples from BAF, the distributions of fuel particles over their masses, sizes, and, eventually, activity median aerodynamic diameters (AMADs). In February-March 1988, we determined the corresponding distributions for 12 settlements of the ChNPP zone. Five of them are placed on the first trace at distances from 8 to 30 km to the west from ChNPP.

The photoradiometric method of measurement of the sizes of fuel particles is based on the analysis of radiographs formed by the coordinate-dependent imprints of the beta-emission of samples from BAF on X-ray films. To create a coordinate system, we hammer nails in three corners of a sample. On them, we put an X-ray film in order to obtain a radiograph. During the exposure in order to conserve the active layer, the sample surface was covered by a thin film, which was practically transparent for the beta-emission and nontransparent for the alphaemission from the sample surface. The duration of the first exposure was 20 min. The durations of the following ones were 1, 3, 9, 27, and 81 h, i. e., each subsequent exposure was thrice longer than the previous one. After the development of a film under the condition of identical illumination, we saw the characteristic point-like dark areas located above the fuel particles, which were well fixed at the examination of a radiograph. The comparison of the imprints of fuel particles on the subsequent radiographs ensured the elimination of false imprints and the confident corroboration of the existence of "true" ones on the previous radiograph.

The essence of PRM consists in the use of each radiograph as a register of fuel particles, whose beta-emission exceeds some threshold value characteristic of the given exposure time sufficient for the detection of the minimally noticeable imprint of a fuel particle on the radiograph.

The input prerequisite assumes that all fuel particles have ball-like shape, and their radionuclidic composition and specific activity correspond to those in Table 1.

The counting rates of beta-particles with $E\beta > 1{,}020$ keV from fuel particles caused the appearance of imprints on 20-min radiographs on all twelve samples from BAF were measured with a spectrometric beta-emission detector collimated down to 5 mm. The measurements were carried out in January 1988. At that time in the chosen energy range, practically only $^{144}Ce = {}^{144}Pr$ (Eg = 2,996 keV), $^{106}Ru = {}^{106}Rh$ (Eg = 3,530 keV), and $^{90}Sr = {}^{90}Y$ (Eg = 2,273 keV) from "nonvolatile" rns gave contributions, whose ratios are presented in Table 1. The accepted assumptions imply that the counting rates for the beta-emission of "nonvolatile" rns for $E\beta > 1{,}020$ keV are proportional to the mass, volume, and, respectively, cube of the "cerium" radius of measured fuel particles. In such way, we got the value of the threshold "cerium" radius for 20-min radiographs, which is equal to 5.34 μm and corresponds to interval 6 in Tables 4 and 5. The threshold values for intervals 2, 3, 4, and 5 were obtained from the ratios of the exposure times in them to that for interval 6. The count-

Table 4. Characteristics of the aerosols of fuel particles settled down on the samples from BAF

Interval		1	2	3	4	5	6	7	8	9	10
r	r_{from}	1.23	1.78	2.56	3.70	5.34	7.71	11.13	16.07	23.2	33.5
	r_{to}	0	1.23	1.78	2.56	3.70	5.34	7.71	11.13	16.07	23.2
$\check{r}$		0.78	1.48	2.13	3.06	4.41	6.34	9.12	13.1	19.8	27.0
V		1.95	13.6	40.5	120	359	1067	3177	9417	27833	82260
AD		5.0	9.55	13.7	19.7	28.4	40.9	58.8	84.5	121	170
^{144}Ce		0.35	2.5	7.3	21.7	65.0	193	575	1704	5037	14886

Note. r, μm — the value of the effective cerium radius of fuel particles at the upper (r_{from}) and lower (r_{to}) boundaries of the given interval of sizes. The values of r are calculated with regard for the fact that the mean specific activity of ^{144m}Ce in a fuel as of 26.04.1986 was $1.74 \cdot 1{,}010$ Bq g^{-1} at a density of 10.4 g cm^{-3}; $\check{r}$, μm — the mean geometric value of the effective cerium radius for the given interval of sizes of fuel particles; $\check{r} = \sqrt{r_{from} \cdot r_{to}}$ for $r_{from} \geq 1.23$ μm; $V = \frac{4}{3} \cdot \pi \cdot (\check{r})^3$, μm^3 — the mean geometric volume of fuel particles in the given interval of sizes; $AD = 2 \cdot \check{r} \cdot \sqrt{\frac{10.4}{1}}$, μm^3 — the aerodynamic diameter of a fuel particle with the given value of $\check{r}$; ^{144}Ce, Bq — the activity of radionuclide ^{144}Ce in a fuel particle with volume V, μm^3, as of 26.04.1986.

Table 5. Characteristics of the aerosols of fuel particles settled down on the samples from BAF taken on the first trace of emergency fallouts from ChNPP as of 26.04.1986

L		Intervals									
		1	2	3	4	5	6	7	8	9	10
30	n		87	106	72	59	12	9	2		
	η	0.02	0.012	0.044	0.089	0.217	0.131	0.293	0.193		
25.4	n		9	29	24	20	6	4	3		
	η	0.01	0.0021	0.0197	0.0485	0.121	0.108	0.214	0.476		
19.6	n		191	107	101	81	18	12	9	2	
	η	0.02	0.0105	0.0172	0.048	0.116	0.0765	0.152	0.338	0.222	
18.5	n		82	87	82	65	28	15	11	1	1
	η	0.01	0.0034	0.0106	0.030	0.070	0.090	0.143	0.312	0.084	0.248
8.1	n		68	25	38	20	20	13	13	1	
	η	0.008	0.0041	0.0044	0.020	0.031	0.093	0.181	0.536	0.122	

Note. L, km — the distance from ChNPP to the sampling place for the given sample from BAF on the trace of the first emergency ejection; n — the number of fuel particles in the given interval of sizes for the given sample from BAF; η, rel. un. — the contribution of the activity of radionuclide ^{144}Ce in the given interval of sizes to the activity of ^{144}Ce in the given sample from BAF.

Table 6. Main characteristics of the fallouts of fuel particles at a distance of L km from the reactor for the samples from BAF and by the results of measurements of the benches on the first trace

L, km,	30	25.4	19.6	18.5	13.8	8.1	4.6
AMAD, μm	41	59	65	70	—	95	—
AD<34 μm	0.382	0.201	0.204	0.124	—	0.068	—
m, g km^{-2},	100	230	330	220	150	580	2,100

Note. AMAD, μm – the value calculated for fuel particles settled down on the surface of the given sample from BAF on the basis of the postulated logarithmically normal distribution of the sizes of settled down particles; AD < 34 μm — the contribution of the activity of ^{144m}Ce in fuel particles characterized by AD < 34 μm to the total activity of ^{144m}Ce in the given sample from BAF; m, g km^{-2} — the fallout density of fuel particles at the sampling place for a sample from BAF at the given distance from ChNPP.

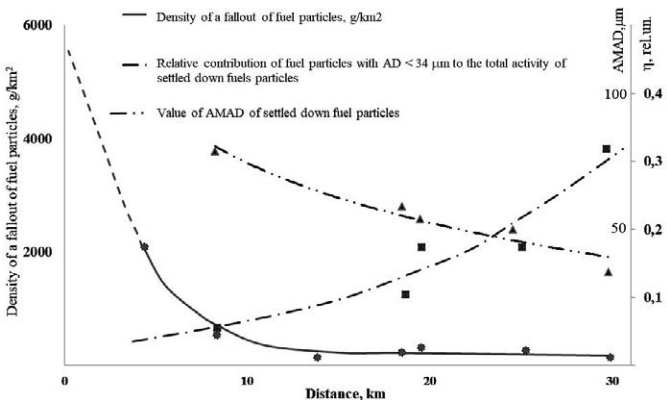


Fig. 4. Graphical representation of the data in Table 6

ing rates exceeding the upper level for interval 6 were distributed over intervals 7, 8, 9, and 10. The 81-h radiographs were used only for the correction of 27-h radiographs. The contributions of the activity for intervals 1 were determined, taking into account that the integral shape of the logarithmically normal distribution of the activity of fuel particles in the probabilistic-logarithmic coordinates should be a straight line.

According to the power production distribution over the active zone at the accident, ~25 % of fuel assemblies were characterized by the specific activity of radionuclide ^{144}Ce less than 1.74 · 1,010 Bq g^{-1}. Thus, the executed evaluation of AMAD in the primary ejection of fuel particles by their activities is the minimum one.

In Fig. 4, we show the data presented in Table 6.

The results of studies of fallouts indicate that the sizes of fuel particles in the primary ejection were essentially larger than at AMAD = 15 μm.

Inhalation entrances

Totally, more than one hundred persons, being present in premises of ChNPP and near them at the time moment of the accident, were delivered to the Hospital no. 6 at once after the accident. For 3 months of their stay at the hospital, 25 persons died: three persons in 86, 91, and 96 days after the accident, and 22 persons in the time period from 14 till 34 days [6].

In Tables 7 and 8, we give the results of processing of the data of measurements on the persons died in the spring and summer of 1986. Tables 7 and 8 present, respectively, the data on the persons, who were present at the time moment of the accident in the NPP premises and outside them (in the first turn, the firemen).

The data presented in Tables 7 and 8 do not show any clear causes, which would lead to the very remarkable differences in the lifetimes of the group of 3 persons (marked by *) after the accident and 22 rest ones. At the same time, it seems obvious that such significant difference is a consequence of some important causes. Therefore, in what follows, we will consider meanwhile only the group of 22 persons.

As of the time moment of the death, the activities of rns deposited in lungs were determined by means of the measurement of the activities of rns in the samples of lung tissues on a PPD gamma-spectrometer. From the lungs of each person, 15 point samples were taken by a definite scheme, for which the specific activities of deposited rns were measured. The ratios in Tables 7 and 8 were obtained by the averaging of measured point values of the specific activity in the lungs of each person. The masses of lungs were accepted to be 1,400 g.

In essence, the values of the activity of fuel particles deposited in the alveolar domain of lungs (respiratory fraction) at the death time were determined in such way. The reduction to the entrance time was carried out by the decay and in correspondence with the cleaning of the alveolar domain according to [12] (see Fig. 5).

The intervals of values of the survival time, τ = (14÷32) days for persons, being outside the NPP premises at the time moment of the accident and τ = (15÷22) days for persons, being in the NPP premises, are typical of the intestinal form of ARS [13].

With the highest confidence, this diagnosis can be referred to the suffered persons marked in Table 8 and Fig. 6 with Nos. 1, 3, 4, 6, 7, 9, and 10 [τ = (14÷20) days] and in Table 7 and Fig. 7 with Nos. 1, 3, 5, 6, 7, 8, and 11 [τ = (15÷21) days].

In this case, the interval of measured values of the dose D was (11÷14) Sv for persons outside the NPP premises with the average value = 12.1 Sv and the median ^{m}D = 12.1 Sv, whereas = 7.4 Sv and ^{m}D = 6.0 Sv for persons in the NPP premises.

The comparison of the doses and the lifetimes indicates that, at the same lifetime after the accident, the values of the dose by hematology, D, received in the NPP premises are approximately by a factor of 1.5 less than the values of the dose D obtained outside the NPP premises.

Of course, by dealing with the results of measurements of the samples of the biological origin, we should remember always that the "use of the results of measurements with indefinite accuracy can become frequently a source of empiric misinformation" (unknown author).

In Table 9, we present the average and median values of the distributions by the data in Tables 7 and 8 and

Table 7. Data on the persons, being in the NPP premises during the accident

No.	Initials	τ, days	*D*, Gy	^{144}Ce, 10^4 Bq	$\frac{^{137}Cs}{^{134}Cs}$	$\frac{^{103}Ru}{^{106}Ru}$	$\frac{^{106}Ru}{^{144}Ce}$	$\frac{^{137}Cs}{^{144}Ce}$	$\frac{^{95}Zr}{^{144}Ce}$	$\frac{^{141}Ce}{^{144}Ce}$	$\frac{^{131}I}{^{137}Cs}$
1	AAF	16	8.9	3.8	2.46	4.9	0.72	0.15	1.83	1.69	—
2	BAI	24	9.3	0.38	5.7	8.9	0.38	0.12	1.72	1.87	67
3	BVS	18	11.0	2.4	1.92	4.0	0.69	0.20	1.88	1.63	15
—	VYuA*	86	7.5	3.2	3.33	4.2	0.41	0.065	1.19	2.02	—
4	DVM	23	1.9	20.4	2.35	7.5	0.56	0.95	2.0	1.87	6
5	KAG	18	4.7	12.0	2.15	4.53	0.53	0.069	1.72	1.49	16
6	KAKh	16	4.7	76.0	2.47	5.76	0.46	0.34	1.94	1.78	3.5
7	LVI	21	5.5	11.0	1.7	4.49	0.56	0.059	1.88	1.56	31
—	NAV*	91	10.1	1.7	2.46	5.23	0.45	0.056	1.35	1.67	—
8	PGI	17	6.4	0.43	2.63	1.44	1.83	0.33	0.90	0.74	—
9	PVV	21	6.1	2.8	2.41	4.73	0.63	0.26	1.64	1.49	17
10	PKG	24	8.2	1.7	2.92	4.29	0.56	0.10	1.81	1.57	—
11	TLF	18	11.8	3.1	1.68	3.28	0.78	0.13	1.72	1.50	11

Table 8. Data on the persons, being outside the NPP premises during the accident

No.	Initials	τ, days	*D*, Gy	^{144}Ce, 10^4 Bq	$\frac{^{137}Cs}{^{134}Cs}$	$\frac{^{103}Ru}{^{106}Ru}$	$\frac{^{106}Ru}{^{144}Ce}$	$\frac{^{137}Cs}{^{144}Ce}$	$\frac{^{95}Zr}{^{144}Ce}$	$\frac{^{141}Ce}{^{144}Ce}$	$\frac{^{131}I}{^{137}Cs}$
1	VNV	19	12.1	0.088	3.46	5.0	1.27	0.52	2.1	1.70	18
2	IEA	30	8.3	0.43	3.6	3.7	0.13	0.13	1.5	1.46	—
3	IVI	17	12.0	0.081	2.46	2.8	0.73	0.73	1.5	1.44	19
4	KVN	15	11.1	0.059	3.56	3.61	4.0	1.48	—	—	33
5	KYuI	32	6.7	1.07	1.93	4.53	0.72	0.1	1.75	1.59	—
—	LKI*	96	4.1	1.01	3.33	5.74	0.25	0.05	1.34	1.77	—
6	OIL	17	12.7	0.58	2.61	3.46	0.87	0.13	1.59	1.38	16
7	PVP	15	13.7	0.41	2.71	4.22	0.73	0.63	1.78	1.59	10
8	SVI	25	6.6	0.106	1.2	9.9	0.37	0.37	1.48	2.72	—
9	NVI	14	10.9	0.052	2.47	—	0.53	0.21	—	—	12
10	TNN	20	12.5	0.21	3.15	5.75	0.46	0.20	1.08	—	35
11	SAA	34	4.4	0.046	2.2	—	—	0.087	—	1.67	—

Note. τ, days — the lifetime after the accident; D, Gy — the personal absorbed dose determined from the blood samples taken and measured at the hospital in several days after the accident; 10^4 ^{144}Ce, Bq — the measured activity of ^{144m}Ce in lungs reduced by its decay and by the excretion (by the data in Fig. 1) to the time moment of the accident, i. e., the activity of ^{144m}Ce in the respiratory fraction of fuel particles; $\frac{Cs^{137}}{Cs^{134}}$ and so on — the values of the typical ratios of the activities of rns deposited in lungs; the activities are recalculated to the time moment of the accident by their decays.

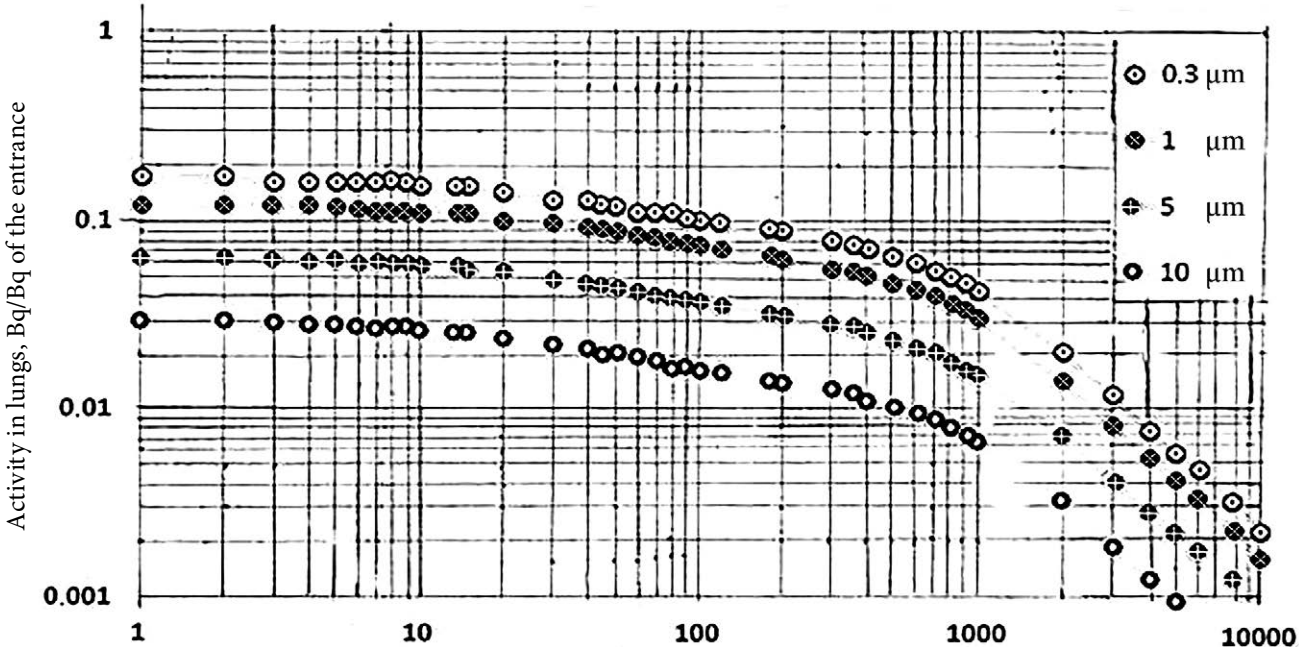


Fig. 5. Time dependence of the activity of long-lived radionuclides in lungs after a single entrance of insoluble aerosols (the M type of joining at the inhalation) for various values of AD [12]

Table 9. Radionuclidic characteristics of the aerosol distributions

Table	^{144}Ce, 10^4 Bq		$\frac{^{137}\text{Cs}}{^{134}\text{Cs}}$		$\frac{^{103}\text{Ru}}{^{106}\text{Ru}}$		$\frac{^{106}\text{Ru}}{^{144}\text{Ce}}$		$\frac{^{137}\text{Cs}}{^{144}\text{Ce}}$		$\frac{^{95}\text{Zr}}{^{144}\text{Ce}}$		$\frac{^{141}\text{Ce}}{^{144}\text{Ce}}$		$\frac{^{131}\text{I}}{^{137}\text{Cs}}$	
	$q_{av}\Delta$	q_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m	$r_{av}\Delta$	r_m
7	10.918	3.3	2.630.99	2.4	4.91.8	4.5	0.660.36	0.56	0.220.24	0.16	1.660.31	1.7	1.610.30	1.56	2119	17
8	0.350.35	0.1	2.720.70	2.5	4.82.0	4.5	0.911.0	0.6	0.390.39	0.2	1.570.27	1.5	1.70.38	1.59	209	18
1			1.90		3.87		0.31		0.064		1.4		1.4		8.8	

Note. q_{av} and Δ — the average value and MSD of the distributions of the activity of ^{144m}Ce in lungs by the data in Tables 7 and 8; q_m — the median values of the distributions of the activity of ^{144}Ce in lungs by the data in Tables 7 and 8; r_{av} and Δ — the average values and MSD of the corresponding ratios for rns by the data in Tables 7 and 8; r_m — the medians.

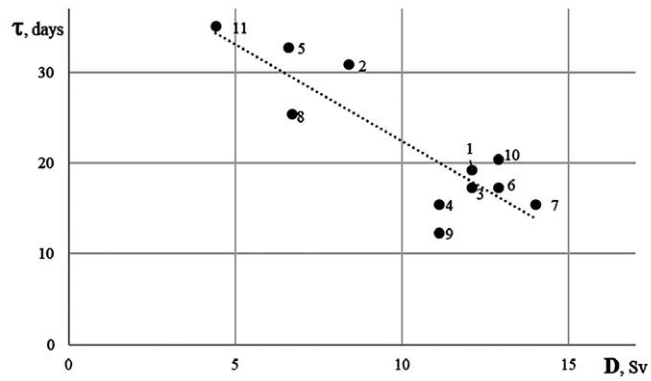


Fig. 6. Regression $\tau = \tau(D)$ drawn by the data in Table 8 (for persons, being outside of the ChNPP premises)

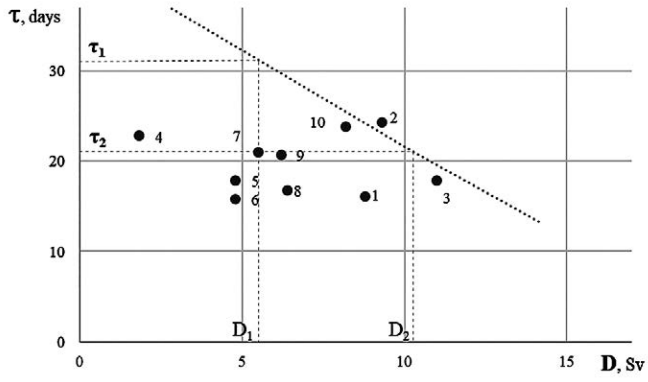


Fig. 7. Graphic representation of the data in Table 7 (for persons, being in the ChNPP premises). The straight line is the regression $\tau = \tau(D)$ from Fig. 6

by the rated data from Table 1 on the activity of radionuclide ^{144}Ce in lungs, as well as the average and median values of the ratios of the activities of rns in the lungs of persons, being inside or outside the NPP premises at the time moment of the accident.

The analysis of the data presented in Table 9 leads to the following conclusions:

on the whole, the values of the radionuclidic ratios $\frac{^{137}Cs}{^{134}Cs}$, $\frac{^{95}Zr}{^{144}Ce}$, and $\frac{^{141}Ce}{^{144}Ce}$ and their variances testify to the sufficiently high reliability of the results of measurements of the activity of rns in the lung samples. Particularly, we emphasize the reliable distinctions of the cesium ratios from the rated fuel ones, which testifies one more to the preferential dispersion of the "young" fuel at the explosion;

radionuclidic compositions of the activity in the lungs of the suffered persons represented in Tables 7 and 8 are identical and are caused by the inhalation of a single aerodispersed system;

values of the activity of rns ^{144}Ce and ^{95}Zr settled down in lungs are completely determined by the inhalation of fuel particles;

the entrance of the radioactivity into lungs of the persons, being in the NPP premises, in the form of fuel particles turned out, on the average, by at least 30 times more than for the persons outside the NPP premises. This circumstance allows us to assume the possibility for the noncontrolled contribution of the inhalation entrance of fuel particles to the values of the dose received by the persons, being in the NPP premises.

Estimation of the internal irradiation dose caused by the entrance of fuel particles

In Table 10, we give the rated values of the dose coefficients for various sections of respiratory organs and gastrointestinal tract (GIT). The results of calculations are presented in the frame of the ideas accepted at the present time [12] that concern the entrance of fuel particles with a logarithmically normal distribution of sizes with AMAD = 10 μm and β_g = 1.5.

In this table, we present the values of the equivalent dose accumulated, respectively, for the first day, first week, and first month after the entrance. The values of the dose are normalized to 1 Bq of ^{144m}Ce in particles of the dispersed fuel with the radionuclidic composition from Table 1.

Specific features of the digestion physiology allow us to think that the doses on digestive organs from the alphaemitting radionuclides contained in fuel particles are negligibly small as compared with those beta- emitting ones in such particles.

The values of the dose coefficients for N are determined by the absence of a standard "physiological" means of the cleaning of basal cavity from entered dust. Such means are: sialophagia for T and mucociliary transport for B and TB, i. e., "automatic" actions without any control by consciousness. But for N, they are the blowing of a nose, sneezing, cough, and expiration, whose efficiencies depend on the fluid secretion intensity determined by the degree of irritation (affection) of sensitive cells of yellow spot, carina, etc. Of course, the preferential deposition of fuel particles with largest sizes and, respectively, their activities must determine the relatively higher value of the

Table 10. Values of the accumulated equivalent dose D for 1, 7, and 30 days after the single entrance in different sections of GIT and respiratory organs caused by the inhalation of aerosols with fuel particles with AMAD = 10 μm in the units 10^{-8} Sv/Bq ^{144}Ce

Days	α, β	stwall	siwall	uliwall	lliwall	colon	et1	et2	bbsec	bbbas	Bb	ai
		S	SI	ULI	LLI	LI	N	T	B		TB	A
1	β	0.55	1.3	5.0	5.1	5.0	3100	8.1	6.0	4.7	1.3	0.36
	α	—	—	—	—	—	350	0.13	1.9	0.32	0.62	0.031
7	β	0.59	1.5	6.5	16	11	4500	11	20	16	4.5	1.7
	α	—	—	—	—	—	560	0.58	9.3	1.8	2.5	0.21
30	β	0.62	1.5	6.6	16	11	4500	13	41	32	9.7	4.7
	α	—	—	—	—	—	560	2.1	26	5.3	6.6	0.75

Note. Days — the time interval after the accident; α and β denote the contributions of alpha- and beta- emitting rns, respectively. S — stomach; SI — small intestine; ULI — upper domain of large intestine; LLI — lower domain of large intestine; LI — calculation for 0.57 ULI + 0.43 · LLI; N — nasal cavity, mouth; T — nasopharynx, throat, larynx; B — bronchioles; TB — trachea and bronchi; A — alveoli.

dose coefficient for N as compared with other parts of respiratory organs, but it is not understandable how large this enhancement will be. So, we restrict ourselves by the assertion that the dose coefficient for N is the largest one.

Fuel particles are not dissolved in fluids of organism. Therefore, the ratios of the dose coefficients for SI and LI are independent of AMAD. The data presented in Table 10 indicate that, due to the inhalation of fuel particles, the accumulated dose on LI is 7.3 times larger than that on SI in 7 days after the entrance.

Small intestine consists of duodenum, jejunum, and ileum, whose total length is 2.3÷3.8 m. The staying time of a substance portion in small intestine is ~4 h. The wall of small intestine is formed by mucous membrane, tela submucosa, and serous membrane. The diffusion and the enzymatic transport of nutrients from the intestine content into fluids of organism occur through mucous membrane [14, 15].

The surface of mucous membrane of SI is characterized by a relief formed by a number of anatomic formations: circular folds, villi, and intestinal glands or crypts.

Having passed SI for 4 h, a substance portion enters LI, which is the end part of human GIT. In LI, there occurs the final absorption and the transfer of simplified substances and water into blood. The undigested unnecessary substances form scybalous masses and leave organism. The length of LI is 0.9÷1.3 m, and the staying time of a substance portion is ~36 h.

Mucous membrane of LI has no villi, but has many folds and a significantly larger number of crypts than in mucous membrane of SI. Their sizes are larger, and their length attains 0.7 mm.

Mucous membrane of intestine undergoes a mechanical action and interacts permanently with the chemically active environment of intestine contents. Under those conditions, the lifetime of functional cells of mucous membrane is less than one week. The constant renewal of the population of functional cells of mucous membrane is ensured by reproducing cells located on the bottom of crypts.

The data of measurements given in Fig. 6 allow to conclude that a single dose of external photonic radiation as high as 12 Sv is absolutely lethal for the population of reproducing cells of mucous membrane namely in small intestine, since even a dose of 20 Gy on large intestine causes serious (hard) consequences only in 5 % of irradiated persons [16].

Tables 7 and 8 contain the measured values of the quantities, whose use allows us to carry out the estimations of individual dosimetric characteristics caused by the inhalation entrance of fuel particles during the first minutes ÷ tens of minutes after the explosion into the persons, who will die later on. Such quantities include the values of the survival time τ [days] and the *in vivo* measured values of the dose by hematology D [Sv].

In Fig. 6, we show the plots of individual values of the dose D in Sv by hematology and the corresponding values of the survival time after the accident by the data in Table 8 for persons, being outside the NPP premises.

Near the points, we presented the numbers from Table 8.

The straight line is the phenomenological dependence $\tau = \tau(D)$ of the lifetime τ on the dose D and is constructed for persons, being outside the NPP premises, i. e., by data in Table 8.

The conclusions of Section 6 imply that, for the persons being outside the NPP premises at the time moment of the accident, the values of the dose D are caused mainly by the external photonic radiation. In the frame of this assumption, we may consider that Fig. 6 gives the dependence of the survival time τ on the external radiation dose.

The attempt to construct a similar regression for the persons, being in the NPP premises, (see Table 7 and Fig. 7) has revealed that, at the same survival time for seven out of eleven died persons, being in the NPP premises, the value of the dose D was essentially (by several times) less than that for the persons, being outside the NPP premises (dose "deficit" Δ). At the same time, the value of the postmortal activity of fuel particles in lungs for the same persons turned out by several tens of times more than that for the persons, being outside the NPP premises.

So, it would be quite reasonable to assume that the dose "deficit" Δ was "filled up" by the internal irradiation caused by the inhalation of fuel particles, and the individual values of the dose "deficit" are the values of the dose on small intestine (Δ_{SI}) caused by the inhalation of fuel particles.

In Fig. 7 in the same format as in Fig. 6, we present the data in Table 7, i. e., for the persons, being in the NPP premises, and the straight line is taken from Fig. 6. In Fig. 7, we give an example how to get the values of $\Delta\tau$ and Δ by data on the suffered person marked by No. 7 in Table 7. Here, $\Delta\tau = \tau_2 - \tau_1 = 10$ days, and $\Delta_{SI} = D_2 - D_1 = 4.8$ Sv.

The data presented in Fig. 7 indicate that, for all suffered persons represented in Table 7, except for No. 2 No. 11, the lifetime corresponding to the given dose is less by $\Delta\tau$ than that following from the dependence $\tau = \tau(D)$. In addition, the dose corresponding to the given lifetime is less by Δ_{SI} than that from the dependence $\tau = \tau(D)$.

The results of the joint processing of the data in Table 7 and Fig. 7 for the persons, being in the NPP premises, are given in Table 11.

Table 11. The results of the joint processing of the data for the persons, being in the NPP premises

No.	1	2	3	4	5	6	7	8	9	10	11
τ, days	16	24	18	23	18	16	21	17	21	24	18
D, Sv	8.9	9.3	11.0	1.9	4.7	4.7	5.5	6.4	6.1	8.2	11.8
Δτ, days	8	0	2	—	14	17	10	12	9	1	0
Δ_{SI}, Sv	3.8	0	0.8	7.5	7.0	8.0	4.8	5.7	4.3	0.7	0
Π, 10^8 Bq	2.5	0	0.5	5.0	4.7	5.3	3.2	3.8	2.9	0.5	0
Δ_{LI}, Sv	27.5	0	5.5	55	52	58	35	42	32	5.5	0
D+Δ_{SI}, Sv	12.7	9.3	11.8	9.4	11.7	12.7	10.3	12.1	10.4	8.9	11.8
D+Δ_{LI}, Sv	36	9.3	16.5	57	57	63	42	48	38	14	12
D_T, Sv	40	9.3	16	65	63	71	46	54	42	14	11.8
Π/L	6,600	—	2,100	2,100	3,900	700	2,900	8,800	10,000	2,900	—
AMAD, μm	200	—	120	120	165	75	140	~300	~300	140	—

Note. No. — number of a suffered person in Table 7; D, Sv – dose by hematology; Δτ, days — "decrease" in the survival time from Fig. 7; Δ_{SI}, Sv — dose "deficit" from Fig. 7; Π, Bq – activity of rn ^{144}Ce under the inhalation entrance of fuel particles into a given person; Δ_{LI}, Sv — dose on LI caused by the inhalation entrance; D+Δ_{SI}, Sv — total dose on domain SI; D+Δ_{LI}, Sv — total dose on LI; DT, Sv — total dose on domain T; Π/L — ratio of deposited rns in upper respiratory paths to that in the alveolar domain calculated by the data in Table 11 and Table 7; AMAD, μm — values of AMAD corresponding to the values of Π/L by Table 12.

In Fig. 8, we present the basic dependences of the deposition of aerosols on various sections of respiratory organs as functions of AMAD [12]. The same data underlie the results of calculations executed in correspondence with [3] and given in Table 12.

The data given in Fig. 8 imply that the values of the dose coefficients for domain T of respiratory organs taken for AMAD = 10 μm remain practically constant for fuel particles with sizes exceeding AD = 3 μm.

With regard for the results presented in Table 10 for 7 days after incident, the individual entrance of the activity of fuel particles is $\Pi_i = \frac{(\Delta_{SI})_i}{1.5 \cdot 10^{-8}}$ (^{144}Ce, Bq); the dose on LI for the given suffered person (D+Δ_{LI}) $i = D_i + \Pi_i \cdot 11 \cdot 10^{-8}$ Sv, and the dose on domain T of respiratory organs $(DT)_i = D_i + \Pi_i \cdot (13 + 2.1) \cdot 10^{-8}$ Sv for the suffered person "i" in Table 7.

In this case, while making conclusions on the basis of the use of the individual values of measured quantities, we should not forget that their values were subjected to the action of many factors, which cannot be accounted. For example, for the dose on LI, it would be useful to know:

distribution of the activity of fuel particles over the substance bulk in LI;

data on the mucus layer on the surface of LI;

Did the person drink water or not?;

the last food intake time;

Did the person take supper with alcohol or without it?;

Did the person use the means of individual protection during the accident or not?

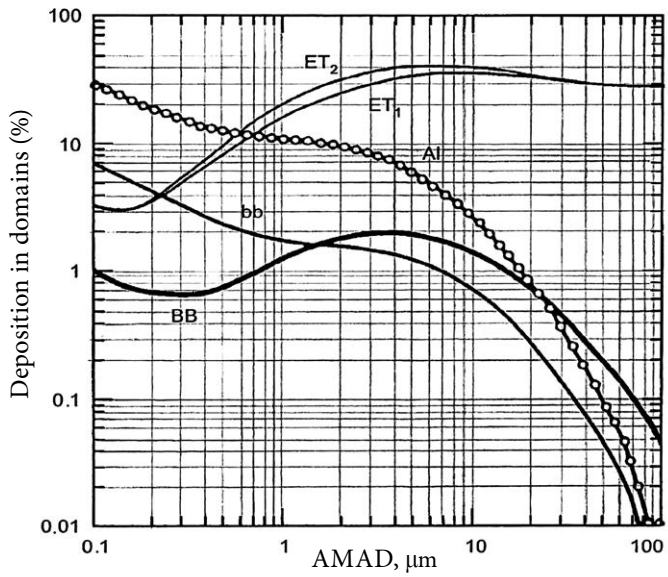


Fig. 8. Dependences of the deposition of aerosols in various domains of respiratory organs on the AMAD of particles [12]

Table 12. Ratio of the activities of particles, which had a uniform distribution of the activity over volume and settled down in upper domains of the respiratory tract and in the alveolar domain of lungs, as a function of AMAD [3]

AMAD, μm	Π/L = (ET2 + BB + bb)/AI
200	7,152
100	1,593
50.0	287
20	48
10	18.6
5	8
1	1.8

It seems that the single thing that cannot happen is as follows: at comparatively small values of the quantity $10^4 \cdot {}^{144}Ce$, no essential dose "deficit" Δ_{SI} can be observed. In this meaning, the positions of points with Nos. 2, 3, 10, and 11 from Table 7 and in Fig. 7 are mainly located properly, i. e., they lie practically on the regression curve.

Generally speaking, there exists the "immense distance" between the rated evaluations of possible values of the internal irradiation dose caused by the inhalation entrance of fuel particles after the explosion and the results of measurements of persons. It is clear that the dose estimations based on "human" data are closer to the "truth", and the studies of fallouts are necessary in order to estimate the causes and the consequences that have defined the fate of the suffered persons. In this aspect, it is necessary to estimate, together with the inhalation of fuel particles, the possible contribution of the inhalation entrance of "hot" particles and "free" atoms to the internal irradiation dose.

Evaluation of the internal irradiation dose caused by the entrance of "hot" particles

The phenomenon of "hot" particles, as possible essential factor of radiation threat, was introduced by researchers from L. Ya. Karpov Institute of Physical Chemistry in the summer of 1986, while studying the aerosols on the breakup of the fourth block.

A particular interest in the activity of rns ^{103}Ru and ^{106}Ru in emergency fallouts arose at the beginning of the summer of 1987, when a hot weather was established. In the measurements of the activity of aerosol filters exposed above the reactor breakup, an anomalous increase in the bulk activity of rn ^{106}Ru in air was detected. Relative to the last winter of 1986–1987, the bulk activity of rn ^{106}Ru increased by tens of times. Simultaneously, the Chornobyl cough "epidemic", which weakened in winter, became again strong. Generally speaking, we cannot assert that there were no attempts in the summer of 1986 to explain the cause of cough. But the hypotheses bore the speculative character and should not be remembered now. All were coughing. I started to cough in August 1986 at a hot weather, when dealt with the measurements of the counting rate of alpha-particles above the surface of soil on streets of the town of Chornobyl with the help of a handheld alpha-radiometer and is coughing till now.

By comparing these circumstances, it turned out that the explanation is quite simple. Ruthenium has oxide with the temperatures of melting, boiling, and decomposition to be, respectively, +25.5 °C, +27 °C, and +108 °C [17]. Thus, the ruthenium anomalies measured on May 15–16, 1986 in the Berezinskii biosphere reserve and in Finland on May 10–12, 1986 [18, 19] were explained. However, it remained unclear how and when ruthenium came out from the fuel matrix and formed .

In the daytime on 27.04.1986, the primary emergency ejection came to Sweden. The measurements of the activity and the radionuclidic composition of aerodispersed samples taken on 27.04.1986 had shown the presence, together with fuel particles, atoms, and molecular compounds of radionuclidic "volatile" elements (iodine, cesium, tellurium, etc.) [20], of "hot" particles of radioactive aerosols on the filters, whose activity was mainly determined by rns $^{106}Ru = {}^{106}Rh$ and ^{103}Ru, which had the rated reactor ratio at the time moment of the accident. The studies of "hot" particles on an electron microscope had shown that they have drop – like shape. The majority of "hot" particles were ~1 μm in diameter. But particles with essentially larger sizes were observed as well: the corresponding photo in [21] represents two typical particles with spherical shape 2.5 and 3.3 μm in diameter. In Table 13, we present the radionuclidic characteristics, which were corrected by us, of three other particles, as an example. We note that the original publication [21] mentioned rn ^{51}Cr, which could not be present in that situation, instead of rn ^{105}Rh and confused rns ^{141}Ce and ^{144}Ce. For particle No. 3, we made calculation and added the result into Table 13 for rn ^{103}Ru. We note that the last column in Table 13 gives the results of measurements of a single "hot" barium particle.

The absence of more complete data on barium "hot" particles is caused, apparently, by the disappearance of the

Table 13. The results of measurements of the activity of three "hot" particles taken in Sweden and measured on April 28–30, 1986

Particle number / Radionuclide	1	2	3
^{103}Ru, Bq	10,000	6,900	155*
^{106}Ru, Bq	2,800	1,800	40
^{105}Rh, Bq	1,700*	—	–
^{99}Mo, ^{99m}Tc, Bq	1,600	—	–
^{132}Te, Bq	810	20	–
^{131}I, Bq	70	60	25
^{140}Ba, ^{140}La, Bq	—	—	2,400
^{95}Zr, ^{95}Nb, Bq	—	—	10
^{144}Ce, Bq	—	—	75*
^{141}Ce, Bq	—	—	120*
^{239}Np, Bq	—	—	70

short-lived system of rns ^{140}Ba = ^{140}La due to the radioactive decay prior to the start of special works on the search for and the study of "hot" particles.

In one year at three different places in Sweden (Stockholm, Gävle, Gotland), the careful seeking of "hot" particles of the Chornobyl origin was carried out on definite areas of the land. As a result, 23 particles were found. Their activities were practically fully determined by rns ^{106}Ru = ^{106}Rh and ^{103}Ru (ruthenium particles) and corresponded to their rated reactor ratio. In Table 14, the results of measurements of five ruthenium particles are presented.

The total specific activity of fuel as of the time moment of the accident was ~1 Bq μm^{-3}. Therefore, it becomes clear from the results of studies given in Table 16 why the ruthenium particles are characterized as "hot" [20].

Studying the element composition of ruthenium particles showed that they "were produced" mainly of molybdenum, ruthenium, and rhodium. At the same places, the Chornobyl fuel particles, whose radionuclidic compositions by their activity correspond to the pre-accident fuel, were found. Moreover, the number of fuel particles detected and collected on the same area was by ~10 times more than the number of ruthenium ones.

This "ratio" agrees with the results of studies of the samples taken in premises of the Shelter object and the samples of soil from the 30-km zone, which were executed at I. V. Kurchatov Institute of Atomic Energy (IAE) in 1987. From the total number of those particles characterized by the activity of at least several tens of Bq, above 90 % were fuel particles. Approximately (4÷7) % were contributed by particles, whose activity at the time of measurements (1988) was due to ^{106}Ru by more than 95 %. As for the radionuclidic composition of fuel particles, their piecewise measurement discovered a "deficit" of cesium. For separate particles, it was as high as 80 % of its cerium ratio averaged over the reactor (see Table 1). On the average, the depletions by cesium and ruthenium were, respectively, ~50 and 20 % [22].

Table 14. The results of studies of five "hot" particles discovered in Sweden in one year after the accident

No.	1	2	3	4	5
Parameters of particles	Haga1	Guan1	Guan2	Haga9	Gävle3
Isotopic composition	Activity, Bq particle^{-1}				
^{103}Ru	17,400	39,300	31,800	3,600	7,900
^{106}Ru	4,350	9,830	9,430	830	1,880
^{125}Sb	3.8	26	15	1.3	—
^{60}Co	1.7	7	3.1	0.1	—
^{144}Ce	—	57	—	—	—
^{137}Cs	0.4	—	—	—	—
Diameter, μm	9.0	11.5	10.7	—	—
AD, μm	29	37	—	—	—
Density, g cm^{-3}	10.3 ± 1	10.7 ± 1	—	—	—
Deposition rate, cm sec^{-1}	2.5	4.2	—	—	—
^{103}Ru, Bq μm^{-3}	49	50	46	—	—

The shape of ruthenium particles, their sizes, and their element composition allowed us to assume that they were formed in the active zone (AZ) during the reactor lifetime from atoms of the metals (fission products) with mass numbers from 95 to 118 (without the 96-th chain). The corresponding sum of values of the yields of those chains per one fission equals 0.51. We verified the validity of the assumption about the origin of "hot" particles. The mean burnout of AZ before the accident was

$$10.9\,\frac{\text{MW day}}{\text{kg(uranium)}} = 9.42\times10^{11} \quad [23].$$

At a power release of 199 MeV/fission, the mean number of fission events (f. e.) was

$$\frac{9.42\cdot10^{11}\cdot10^{-3}}{1.99\cdot10^{2}\cdot1.6\cdot10^{-13}} = 3.0\cdot10^{19}\ \text{f.e. /g uranium.} \qquad (1)$$

At the time moment of the accident, the mean specific content of rn ^{106}Ru in the fuel was 3.7 · 10^{-5} g/g (uranium) [23]. From whence, the mean specific activity of rn ^{106}Ru for λ = 2.15 · 10^{-8} sec^{-1} was

$$3.7\cdot10^{-5}\,\frac{6.02\cdot10^{23}}{106}\,2.15\cdot10^{-8} = 0.45\cdot10^{10}\ \text{Bq/g (uranium);} \qquad (2)$$

for the reactor lifetime, the mean number of fission events per 1 Bq of rn ^{106}Ru was

$$\frac{3.0\cdot10^{19}}{0.45\cdot10^{10}} = 6.7\cdot10^{9}\ \text{f.e./Bq (^{106}Ru).} \qquad (3)$$

Thus, for the mean mass number of fission fragments in the indicated chains M = 99 [24], the mass yield of metals for one fission event as of 26.04.1986 was:

$$\frac{99\cdot0.51}{6.022\cdot10^{23}} = 8.39\cdot10^{-23}\ \text{g/f.e.} \qquad (4)$$

Respectively, the rated value of the mass yield per 1 Bq for rn ^{106}Ru was

$$\theta = 6.7 \cdot 10^{9} \cdot 8.39 \cdot 10^{-23} = 5.6 \cdot 10^{-13} \text{ g/Bq } (^{106}\text{Ru}). \quad (5)$$

On the other hand, the mean value of the mass yield as of 26.04.1986 was calculated by the data of measurements of three particles given in Table 14 (Haga 1, Guen 1, Guen 2) and turned out to be

$$\theta = (8.3 \pm 0.9) \cdot 10^{-13} \text{ g/Bq } (^{106}\text{Ru}). \quad (6)$$

These values (calculated and measured) of the mass yield coincide in the limits of ~15 %, i. e., are sufficiently close to each other. This fact corroborates the soundness of the assumed origin of "hot" particles.

On the microphotos, "hot" particles have a clearly manifested drop-like shape, which testifies that they were formed from a melt and were localized in the intergranular space of fuel pellets at the normal pre-accident operation of the reactor.

The formation of inclusions occurred due to the release of atoms of FPs from the fuel matrix in the central most heated region of fuel pellets during the whole reactor lifetime. In this case, the absence of significant amounts of "volatile" FPs in ruthenium and barium inclusions is caused by the fractioning due to the migration of free atoms to the "cold" periphery of pellets with the subsequent localized "freezing" of elements depending on their melting temperature and the temperature gradient along the fuel pellet radius. It explains the separation of ruthenium and barium and the practical absence of "volatile" FPs in the samples of "hot" particles taken in Sweden.

In Table 15, we present the results of measurements of the activity of rn ^{106}Ru for five particles from Table 14 (Nos. 1, 2, 3, 4, and 5), the activity of rn ^{106}Ru or ^{103}Ru for two particles from Table 13 (Nos. 6 and 7), and the measured diameters (2.5 μm and 3.5 μm) of two particles (Nos. 8 and 9). The rest values of the activity of ruthenium and the diameters of particles represented in Table 14 were calculated for $\theta = (8.3 \pm 0.9) \cdot 10^{-13}$ g/Bq (^{106}Ru) and $\rho = 10.5$ g cm^{-3} as of 26.04.1986.

At the time moment of the accident, the total activities of rns ^{106}Ru and ^{144}Ce in the pre-accident reactor were, respectively, 23.2 and 105 MCi. This implies that the mean ratio of the specific activities of ^{106}Ru and ^{144}Ce in the fuel at the time moment of the accident was 0.22.

Let q_f be the specific activity of rn ^{106}Ru in fuel granules at the time moment of the accident, and let q_{free} be the "free" specific activity of ruthenium released from fuel granules. Then, for the measured values of the specific activities $^{106}\text{Ru}_{meas}$ and $^{144}\text{Ce}_{meas}$, the ratio of free ruthenium and fuel one is

$$r_{Ru} = {}^{106}\text{Ru}q_{free} / {}^{106}\text{Ru}q_f = ({}^{106}\text{Ru}q_{meas} - 0.22 \cdot {}^{144}\text{Ce}q_{meas}) / / 0.22 \cdot {}^{144}\text{Ce}q_{meas}. \quad (7)$$

The normalization of the ^{144m}Ce activity is due to that, as was noted above, refractory cerium is one of the most "nonmobile" elements among FPs in the fuel matrix, and its measured specific activity ${}^{Ce144}q_{meas}$ is a fuel marker.

Analogously, the ratio of the free activity to the fuel one for ^{137m}Cs is

$$r_{Cs} = {}^{137}\text{Cs}q_{free} / {}^{137}\text{Cs}q_f = ({}^{137}\text{Cs}q_{meas} - 0.032 \cdot {}^{144}\text{Ce}q_{meas}) / / 0.032 \cdot {}^{144}\text{Ce}q_{meas}. \quad (8)$$

The calculated ratio of specific activities of ^{137}Cs and ^{144}Ce in a fuel ${}^{137}\text{Cs}q_f / {}^{144}\text{Ce}q_f = 0.064$.

The value $r_{Cs} = 0.032$ was chosen with regard for the depletion of fuel granules with cesium down to 50 % due to the release of cesium from a fuel.

It is seen from the data of Table 16 that, on the western trace formed directly by the reactor explosion fallouts, the contribution of free ruthenium, ${}^{Ru106}q_{free}$, to the measured activity ${}^{Ru106}q_{meas}$ is the least: $r_{Ru} = 0.23$. Along the other directions, the fallouts formed after some time by the aerosols from the reactor breakup have the significantly larger ruthenium ratio: for example, the fallout formed in the SW direction in ~5 days after the explosion had $r_{Ru} = 2.0$, i. e., higher by almost one order.

Table 15. Activities, diameters (d), and aerodynamic diameters (AD) of nine ruthenium particles found and studied in Sweden [20, 21]

No.	1	2	3	4	5	6	7	8	9
^{103}Ru, Bq	17,400	39,300	31,800	3,600	7,900	10,020	6,900	409	934
^{106}Ru, Bq	4,350	9,830	9,430	830	1,880	2,800	1,800	102	234
d, μm	8.7	11.4	11.2	4.98	6.5	7.5	6.4	2.5	3.3
AD, μm	26	34	34	15	19	23	19	7.5	10

Table 16. Ratios r_{Ru} of free ruthenium and fuel one in samples from the BAF taken in different directions from the NPP

Direction	W*	N	NE	E	SE	S	SW
r_{Ru}	0.23	0.93	0.56	0.34	0.56	0.57	2.0
Δ	0.12	0.65	0.05	0.03	0.26	0.09	1.6
n	7	6	4	4	7	5	6

Note. *n* — number of samples for the given direction; Δ — mean square deviation (MSD).

The results of studies of the Chornobyl fallouts in Sweden indicate that, at the time moment of the explosion, the metallic inclusions were released from intergranular cavities of a fuel and formed the fraction of "hot" particles, whose activity was mainly determined by rns ^{106}Ru = ^{106}Rh, ^{103}Ru, and ^{99}Mo and short-lived rns such as, for example, ^{105}Rh, in the aerodispersed system of the primary emergency ejection together with fuel particles and "volatile" atoms and compounds of FPs.

Eventually, we may assert that the formation of volatile from dispersed ruthenium inclusions started only in the fallouts after the explosion that destroyed the reactor.

In Table 17, we give the values of the ruthenium ratio $R_{Ru} = {}^{Ru106}q_{free}/{}^{Ru106}q_f$ calculated by the results of postmortal measurements of the activities of rn ^{144}Ce and ^{106}Ru in the lungs of persons who were present at the time moment of the accident in premises of ChNPP and outside them. According to the data, $R_{Ru} = 2.2 \pm 1.71$ for the persons in premises of NPP and $r_{Ru} = 2.5 \pm 1.6$ for those outside them. The practical coincidence of the ruthenium ratios confirms the conclusion that the radionuclidic compositions of aerosols in premises of NPP and outside them after the accident were identical. The mean value of the ruthenium ratio for both groups is $r_{Ru} = 2.2 \pm 1.5$ with the median value $r_{Ru} = 2.0$.

In organism in the process of inhalation, there occurs the separation of aerosols over sizes due to their gradual deposition on the surface of air-bearing channels of respiratory organs. So, alveoli "are reached" mainly by aerosol particles with AD ~1 μm. In the aerodispersed system formed after the explosion, the ruthenium ratio was at most $r_{Ru} = 0.23$. At the inhalation after the passage of air-bearing channels of respiratory systems of persons by aerosols, it increased up to $r_{Ru} \approx 2.2$, i. e., the relative contribution of the highly dispersed component in alveoli turned out to be ~10 times more as compared with the distribution of fuel particles in the air.

From whence in the "first approximation," we may consider that the distributions of sizes of ruthenium particles in the ejection are characterized by AMAD ≈ 10 μm. This conclusion does not contradict the data in Table 15, since a significant part of the sampling is formed by the

Table 17. The results of in vivo measurements of the absorbed dose by blood samples, postmortal measurements of the activities of rns ^{144}Ce and ^{106}Ru in lungs, specific activity of rn ^{137}Cs in muscles, and the corresponding values of the ruthenium ratio r_{Ru}

In premises of NPP								Outside premises of NPP							
No.	Initials	D_h, Gy	τ,days	^{144}Ce, 10^4 Bq	^{106}Ru, 10^4 Bq	Q,Bq g^{-1}	r_{Ru}	No.	Initials	D_h, Gy	τ, days	^{144}Ce, 10^4 Bq	^{106}Ru, 10^4 Bq	Q,Bq g^{-1}	r_{Ru}
1	AAF	8.9	16	3.3	1.44	2.8	2.1	1	VNV	12.1	19	0.088	0.11	0.7	4.9
2	BAI	9.3	24	0.33	0.15	–	0.79	2	IEA	8.3	30	0.43	0.26	3.6	2.0
3	BVS	11.0	18	2.1	0.86	3.6	2.1	3	IVI	12.0	17	0.081	0.099	0.2	4.8
4	DVM	1.9	23	20.8	13.6	855	1.5	4	KYuI	6.7	32	0.059	0.73	0.9	2.4
5	KAG	4.7	18	10.8	4.24	2.7	1.4	5	OIL	12.7	17	1.07	0.49	0.3	3.0
6	KAKh	4.7	16	66.0	29.9	729	1.1	6	PVP	13.7	15	0.58	0.3	5.4	2.4
7	LVI	5.5	21	9.4	3.22	1.8	1.5	7	SVI	6.6	25	0.41	0.039	0.4	0.77
8	PGI	6.4	17	0.37	0.54	0.7	7.2	8	TVI	10.9	14	0.106	0.02	—	0.05
9	PVV	6.1	21	2.4	1.01	9.6	1.9	9	KVN	11.1	15	0.052	—	0.5	—
10	PKG	8.2	24	1.5	0.89	1.5	1.5	10	TNI	12.5	20	0.21	—	0.6	—
11	TLF	11.8	18	2.7	2.38	0.7	2.6	11	SAA	4.4	34	0.046	—	0.4	—

Note. No. — ordinal number corresponding to Tables 7 and 8; τ, days, — the survival time after the explosion; Dh, Gy, — personal values of the absorbed dose by hematology, which were evaluated in a hospital; ^{144}Ce, 10^4 Bq — activity of rn ^{144}Ce measured in lungs and reduced by decay to 26.04.1986; ^{144}Ru, 10^4 Bq — activity of rn ^{106}Ru measured in lungs and reduced by decay to 26.04.1986; Q(τ), Bq g^{-1} — specific activity of rn ^{137}Cs in the samples of muscular tissue; r_{Ru} — ruthenium ratio $r_{Ru} = {}^{Ru106}q_{free} / {}^{Ru106}q_f$ in lungs of the suffered persons reduced by decay to 26.04.1986.

results of measurements on land in one year after the accident, when the fine ruthenium particles became unnoticeable due to the full decay of rn ^{103}Ru and a significant decrease in the activity of rn ^{106}Ru.

The total partial contribution of the activity of rns ^{99}Mo, ^{103}Ru, and ^{106}Ru = ^{106}Rh calculated by the data in Table 1 to the fuel dose coefficient $K = 1.1 \cdot 10^{-7}$ Sv/(Bq ^{144}Ce) was $\sim 0.2 \cdot 10^{-7}$ Sv/(Bq ^{144}Ce). The partial contribution of free ruthenium was $0.2 \cdot 10 - 7 \cdot 0.23 \approx 0.04 \cdot 10^{-7}$ Sv/(Bq ^{144}Ce), i. e. the contribution of free ruthenium to the internal irradiation dose, which was caused by the inhalation entrance of a dispersed fuel, was less than 5 %.

Dose contribution of "volatile" cesium

As a result of the explosive seal failure and the dispersion of fuel elements, a part of the radionuclides of cesium entered the composition of fuel particles (insoluble cesium), whereas another part turned out in the composition of the "volatile" fraction (soluble cesium).

Cesium is an alkaline metal and, like its chemical analogs (sodium and potassium), is practically uniformly distributed over soft tissues approximately in one day after the inhalation of its soluble form in organism. The appendix of the dissertation by S. I. Dement'ev [6] includes the results of measurements of the specific activity of ^{137m}Cs in the urine samples during the whole time of the stay of suffered persons in the hospital and the results of measurements of the specific activity of rn ^{137}Cs in postmortal samples of muscular tissue.

By the urine samples of all suffered persons (107 persons), being in the hospital, the median value of the effective excretion half – period for soluble cesium was established to be $T_{1/2} = 37$ days, i. e., $\mu_{eff} = 0.019$ day^{-1}.

According to [25], the maximum permissible dose (MPD) is 15 rad yr^{-1} or 0.15 Gy yr^{-1} for soluble ^{137}Cs in the group of tissues II, and the corresponding value of its permissible content (PCA) in muscles is 14 μCi or 0.52 MBq. In other words, for the permanent content of $5.18 \cdot 10^5$ Bq in muscles, the dose per year is 0.15 Gy yr^{-1}. From whence, the dose coefficient is

$$^{137}Cs D = \frac{0.15}{5.18 \cdot 10^5 \cdot 365.25} = 7.93 \cdot 10^{-3} \text{ Gy/Bq} \cdot \text{day}. \quad (9)$$

For the mass of muscular tissue equal to $2.9 \cdot 10^4$ g, the dose from soluble ^{137}Cs for the survival time is

$$^{137}Cs D_{(\tau)} = 7.93 \cdot 10^{-10} \cdot 2.9 \cdot 10^4 \cdot Q_{(\tau)} \cdot e^{\mu_{eff}\tau} \cdot \int_0^{(\tau)} \cdot e^{\mu_{eff}\tau} \cdot dt = \\ = Q_{(\tau)} \cdot 1.21 \cdot 10^{-3} \cdot (e^{\mu_{eff}\tau} - 1) \cdot \text{Gy}, \quad (10)$$

where τ, day, stands for the survival time; and $Q_{(\tau)}$, Bq g^{-1}, is the measured value of the specific activity of ^{137m}Cs in postmortal muscular samples.

On the average in the emergency fallouts as of 26.04.1986, the ratio of ^{137}Cs activity to that of ^{134}Cs was 2.2, and the ratio of the dose coefficient of ^{137}Cs to that of ^{134}Cs was 1.7. From whence, the dose from the rns of cesium is

$$^{Cs}D = {}^{137Cs}D + {}^{134Cs}D = {}^{137Cs}D + {}^{134Cs}D \cdot \frac{1}{2.2 \cdot 1.7} = 1.28 \cdot {}^{134Cs}D. \quad (11)$$

The largest specific activity of ^{137m}Cs in the sample of muscular tissues Q = 855 Bq g^{-1} (No. 4, DVM, in Table 7) at the survival time τ = 23 days corresponds to the highest (among all died persons) dose addition from the entrance of "volatile" cesium:

$$AD = 1.28 \cdot 854 \cdot 1.21 \cdot 10^{-3}(e^{0.019 \cdot 23} - 1) = 0.72 \text{ Gy}. \quad (12)$$

This value of the dose from the internal irradiation caused by the soluble rns of cesium is not low. But, as compared with the dose from the external radiation as high as 1.9 Gy (No. 4, DVM, in Table 7) and from the internal irradiation caused by the inhalation of fuel particles, this addition from soluble cesium had no crucial effect on the fate of this person. The same is true, all the more, for "cesium additions" into organisms of the rest died persons.

Discussion of the results

The values of the dose measured by hematology for eight suffered persons (see Fig. 6) and two persons (Fig. 7) for the insignificant inhalation entrances are in the limits 12–14 Gy, and the survival time for them was 14–20 days. The death of those persons was caused by the definite effect, namely, by the failure of the system of reproduction of the mucous membrane of small intestine at doses exceeding 10–12 Gy. It seems improbable that the external irradiation doses and the induced consequences turned out practically identical for all persons stayed at the time of the explosion and at once after it at different places of the territory and premises of ChNPP. The possible cause consists in that the identical measured doses by hematology were determined by the "saturation" of the biological effect at doses exceeding the aboveindicated value. The real doses of the external radiation can be essentially higher than this level with the same consequences for the suffered persons.

Respectively, the measured doses by hematology, which were at most 8 Gy, can be considered as those cor-

responding to the reality. Then, for the suffered persons Nos. 1, 4, 5, 6, 7, 8, and 9 (see Table 7), the evaluations of the possible contribution to the internal irradiation dose (dose "deficit"), which were executed according to the procedure represented in Fig. 7, are minimum, and the real contribution caused by the inhalation of fuel parvticles to the dose acted on gastroenteric tract and respiratory organs could be significantly higher.

The lifetime of mature functional cells of one-layer cylindrical epithelium is ~3 days. The reproduction of cells occurs by means of the division of cambial and stem cells located at the bottom of intestinal crypts. The "newly born" cells are moving gradually from the crypt bottom to a "destination" approximately for 2–3 weeks. Therefore, the intestinal wall in norm contains simultaneously 5÷6 generations of functional cells of different ages. The death caused by the destruction of the reproducing system occurs with the "wear" of the last generation.

As for suffered persons Nos. 2, 5, 8, and 11 (see Fig. 6), the values of the dose by hematology and the corresponding values of the survival time are determined, the most probably, by specific features of the reproducing system. If the reproducing cells would die completely, the lifetime of persons would hardly turn out more than two weeks. Apparently, the reproducing cells died completely not in all suffered persons, but the vital ability of remaining fragments of the population of those cells turned out lower than the critical value sufficient for the timely reproduction.

The interval of the values of the survival time for doses of (4.4–14) Sv was from 34 down to 14 days and, apparently, was determined by the different numbers of remaining viable fragments of the reproducing system of one-layer cylindrical epithelium in different persons [15].

As for three persons who lived 3 months after the accident, the estimations of the doses on small intestine are as follows:

VYuA	86 days	$D+\Delta f = 13$ Gy;
NAV	91 days	$D+\Delta f = 13$ Gy;
LKI	96 days	$D+\Delta f = 6$ Gy.

By virtue of some individual circumstances, they managed to go through the critical point of a regeneration of the reproducing system of one-layer cylindrical epithelium. But, in 3 months, they were overtaken, apparently, by bone-cerebral syndrome.

At the mean energy of the beta-radiation of fuel particles $\beta \approx 340$ keV (according to Table 1), the path of beta-particles in soft biological tissue is about 1 mm. Thus, the beta-emission of fuel particles that fell in GIT was a significant addition to the external radiation dose acted on the population of reproducing cells of mucous membrane, and the so-called dose "deficit" in the suffered persons stayed in premises of ChNPP was "added" by the inhalation entrance of fuel particles.

In the frame of the conception accepted now [4, 26], it is considered that the internal irradiation of persons caused by the inhalation entrance of the activity at once after the reactor explosion was not able to essentially affect the fate of the persons died from ARS during the first month after the accident.

Generally speaking, the main factors determining the doses onto organs and tissues of the persons subjected to the inhalation entrance of radionuclides (rns) [27] are as follows:

radionuclidic composition of the entrance;

radioactivity of entered rns;

physico-chemical characteristics of the entrance in the gaseous state and in the composition of aerosol particles;

solubility of aerosols in the fluids of organism;

distribution of sizes of aerosol particles (dispersity) characterized by the values of AMADs.

The executed analysis showed that all main factors, except for the dispersity, are practically identical in the traditional approach and in the present work.

It is worth to note that the universal means of description with the use of the logarithmically normal law of a distribution of the sizes of particles in aerodispersed samples is a sufficiently serious idealization. Nevertheless, nothing better is available.

While substantiating the traditional conception, the value AMAD = 15 μm was accepted on the basis of a purely speculative reasoning [4].

At the same time, the results of direct measurements of the dispersion spectrum of fuel particles in thin-layer samples of fallouts given in Fig. 4 indicate that, at distances from 8 km to 30 km along the trace of fallouts of the primary ejection, the values of AMADs decrease uniformly from 95 μm down to 50 μm.

It is worth to note that these measurements are the unique direct measurements of the distributions of the dispersity of fuel particles in the fallouts from the primary emergency ejection near the ChNPP.

Of course, the ratios of the distributions of the sizes of aerosol particles in fallouts and in the air are a sufficiently indefinite thing. But anyway, it seems improbable that the distributions of the sizes of fuel particles after the explosion can be characterized for all suffered persons by the same value of AMAD (15 μm) on the territory and in premises of ChNPP.

In this connection, the estimations of the individual values of AMADs by the data of "human" measurements given in Table 11 do not contradict the studies of five thin-layer samples from BAF taken on the trace of primary fallouts, whose results are presented in Table 5 and in Fig. 4. In any case, all they exceed essentially the value of AMAD = 15 μm accepted in the traditional conception.

The clinic manifestations of the irreversible action of inhaled fuel particles on the GITs of the deceased persons would be established after the anatomic investigations. At the same time, the inhaled fuel particles before their passage into GIT are distributed on the surfaces of various parts of respiratory organs at once after the entrance, which depends on their values of AD. By the estimates presented in Table 11, the corresponding values of the accumulated dose are so high that they would lead to severe oropharyngeal manifestations and to lesions visible "with the naked eye" in the domain G, oral cavity adjacent to it, nasal passages, etc.

More than one hundred persons from those who were at the night on April 26, 1986 in premises and on ChNPP's territory were taken out and hospitalized approximately in two days in the special clinic in Moscow. Thus, the systematic observations and intense therapy were started approximately in two days after the accident. Twenty five persons from them died in the clinic during the spring and summer of 1986.

At the present time, it is considered that the main dose-forming factor was the external gamma- and beta- radiations at accident's night. However, the retrospective evaluations of the external irradiation dose by the data of hematological measurements do not correspond in a number of cases to the heaviness of consequences and cannot serve the explanation of a death under conditions of the clinic.

The generalized clinic data on 107 patients are presented in book [26].

In the third-fourth day after the accident, 80 patients manifested the signs of oropharyngeal syndrome. In the tenth day, the initial manifestations developed to radiation-induced mucositis in various forms and of various heavinesses up to the third or fourth degree.

I would like to present some citations from [26] (pp. 42–45):

"It is necessary to indicate a somewhat higher frequency of mucositis as compared with patients involved in other accidents…";

"Intestinal syndrome (IS) was one of the most severe threatening manifestations of ARS. Its development in ten patients after the short-time irradiation in doses approaching 10 Gy was noticed in 4–8 days…";

"The manifestations of IS were combined simultaneously with ulcerative stomatitis with the discharge of a great amount of mucus containing sometimes blood. These violations hampered not only the food intake, but also the respiration of patients, since they touched the entry to larynx…";

"Acute radiation pulmonitis (earlier observed at the therapeutic gamma-irradiation of patients suffered from leucosis) occurred in seven patients suffered from ARS of the IV degree… The disease was terminated by the development of hypoxic coma in 14–30 days after the irradiation. As a rule, those manifestations were combined with signs of radiation-induced damages of skin and intestine…";

"As for the earlier period, the action of emissions was recognized to be defining in pathogenesis. In the subsequent analysis, the assumption about a possible omission of the action of various chemical toxic factors (products of the combustion of plastics and other structural materials, gas-and aerosol-like ejections of the fuel, and materials thrown in the reactor for the combustion stoppage and the restriction of ejections) was advanced…";

"The dosimetric studies of the contents of cesium and iodine in organism were carried out directly in hospital rooms and, while extending the mode, at a laboratory on the whole-body counters… Those qualified measurements confirmed the extreme rarity of the significant incorporation of radionuclides and the dominant significance of external radiation for the development of changes in the health state of suffered persons" [26] (pp. 47–48).

It is obvious that some understandable gaps in the comprehension of the situation forced the authors of book [26] "to include" products of the combustion of plastics into the causes of lethal consequences. Moreover, no materials were thrown into the reactor at the night of the explosion, and the "gas- and aerosol-like ejections of the fuel" were, first of all, the cause for the incorporation of radioactivity by means of the inhalation and for the irradiation of upper respiratory paths and intestine. Thus, we are faced with the contradiction between the accepted evaluations of the dose and its clinic manifestations. This is especially significant for the part of suffered persons, who were in ChNPP's premises at the time moment of the accident.

All becomes clear if we take into account that the analysis in [26] did not include the evaluation of the dose on GIT in the first day. This dose was determined by the entrance of the inhalated coarse-dispersive aerosol component of the dispersed fuel and by its transit from upper respiratory paths through GIT.

At first glance, it seems paradoxical that the inhalation entrance can cause the lethal dose on small intestine. But let us remember that the case in hand involves insoluble aerosols formed at the explosion of the active zone contained the spent reactor fuel. Those aerosols from the destroyed reactor were "pulled" by a "draught" and by a regular ventilation through the working premises, where the night shift of ChNPP's workers was present.

As usual, the evaluation of the doses on the upper parts of respiratory organs and on GIT is carried on by the results of measurements of the radioactivity of insoluble aerosols that passed by the transit in the first day after the deposition through GIT into excrements, whose samples should be taken during the first days after the entrance of aerosols.

It is probable that, at the hospitalization of suffered persons, the sampling of excrements for the measurements was not performed for the first several days, and the "…qualified measurements … on the whole-body counter" were executed considerably later than the transit purification of upper respiratory paths through GIT was terminated, i. e., "they have missed the train". Thus, one of the significant dose-forming factors which had led to severe consequences was not noticed.

Conclusions

1. On the basis of an analysis of the results of own studies of the emergency fallouts, as well as the results of clinic observations performed at the Hospital no. 6 in Moscow, the presence of a dose contribution from the transit of fuel particles is established, and its estimation for those persons, who was in premises of ChNPP at the emergency time and died later on, is carried out. It is shown that the cause of their death can be the intestinal form of acute radiation sickness arisen to the joint action of the external photonic irradiation and the internal beta-emission radionuclides on the system of reproduction of one-layer cylindrical epithelium of small intestine at the transit of inhalated fuel particles through GIT.

2. The analysis of the totality of formed circumstances allowed us to consider the inhalation of radionuclides ^{103}Ru and ^{106}Ru in the composition of volatile molecules RuO4 formed in the fallouts of ruthenium "hot" aerosol particles as the cause of the "Chornobyl cough" widely spread in the summers of 1986 and 1987 on the territories subjected to the intense fallouts from ChNPP.

3. At the timely detection of the transit of radioactive substances through GIT, the doses of irradiation of GIT and parts N and G of respiratory organs can be decreased by means of the immediate washing-out. Respectively, on the enterprises where there exists a potential danger of the emergency entrance of radioactive aerosols, the facilities of measurements aimed at the rapid detection of the entrance of radioactive substances in human organism and the regulations ensuring, if necessary, the timely decision-making concerning the urgent realization of protective measures must be foreseen.

4. As for the analysis of consequences of the accident at ChNPP, it is expedient to continue the work by means of the comparison of the obtained estimates with the clinic manifestations revealed and registered in the spring and summer of 1986 in the persons, who were hospitalized at once after the accident.

Afterward

For me, this work was started at the end of July 1986, when I was sent to the official trip to the town of Chornobyl and continues till now with the interruption from 1997 till 2016. Nobody ordered this work to me. The impulsive cause was, in particular, the desire to find the origin of a mass cough in the zone of emergency fallouts near ChNPP in the summers of 1986 and 1987. The present work is based on the results obtained by my request and with my participation. I thank sincerely the essential help and the fruitful cooperation with my following colleagues.

S. Yu. Antropov processed the results of applications of the photoradiometric method to the measurement of samples from BAF on a computer in 1989.

V. N. Danilenko measured samples from BAF on a PPD gamma-spectrometer in 1988.

S. I. Dement'ev measured the postmortal samples of tissues of the persons died in the spring and summer of 1986 and the samples of urine and blood of all persons hospitalized at once after the accident on a PPD gamma-spectrometer.

A. A. Molokanov executed the dosimetric calculations in the frame of the respiratory models of ICRP in 2016–2017.

S. V. Petrov participated in field measurements and in the processing of their results in the summer and autumn of 1986.

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The time runs, and some persons passed away. A. K. Gus'kova and I. B. Keirim-Markus are among them. I am grateful to them for the support and the interest in the above-presented results. As before, they are with me.

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Паливні частинки в наслідках аварії на Чорнобильській АЕС

У результаті ядерного вибуху на четвертому енергоблоці Чорнобильської АЕС у повітрі над її територією з'явилася радіоактивна хмара, що містила аеродисперсну систему з аерозолями, що утворилися під час вибуху. Аварія сталася в кінці реакторної кампанії ще до передбачуваного перезавантаження активної зони. Таким чином, до складу хмари входили продукти поділу й активації урану, які накопичувалися в паливі реактора під час кампанії. У ту ніч східний вітер ніс радіоактивну хмару на захід, залишаючи аерозольні випадання на поверхні Землі як радіоактивний слід. Представлено результати власних досліджень (1986–1990 рр.) аерозольного випадання на західному сліді, що утворився відразу після вибуху на Чорнобильській АЕС. Виходячи з цього, реконструюються характеристики (фізико-хімічні форми, радіонуклідний склад, медіанний аеродинамічний діаметр активних частинок (АМАД) і т. п.) аеродисперсної системи, створеної відразу після вибуху активної зони реактора. У рамках респіраторної моделі, наведеної в публікації МКЧХ 66, оцінюються внески, спричинені вдиханням частинок палива (мікроскопічних частинок, які є фрагментами паливних елементів, що вибухнули, і мають в основному їхні радіонуклідні характеристики), в дози опромінення частин органів дихання і шлунково-кишкового тракту. Показано, що причиною масового кашлю влітку 1986 і 1987 рр. на територіях, що зазнали дії аварійних випадів, стало вдихання радіонуклідів рутенію у вигляді , що утворилися в "гарячих" частинках на повітрі і потім випаровувалися з них. "Гарячі" частинки — це компактні включення, утворені продуктами поділу. Вони складаються в основному з атомів, близьких до благородних металів (молібдену, рутенію, родію тощо), що утворюються в звичайний робочий період у паливних таблетках і вивільняються з останніх під час вибуху активної зони. Пояснюються причини неузгодження між клінічнимии наслідками та значеннями доз, що приписані потерпілим, які під час вибуху перебували в приміщеннях АЕС, а потім померли від гострої променевої хвороби через три-чотири тижні після аварії.

Ключові слова: Чорнобильська АЕС, аварія, ядерне паливо, частинки палива, "гарячі" частинки, "летюча" фракція, гостра променева хвороба.

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