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Original Article

Study of the Effect of $(U_{0.8}Pu_{0.2})O_2$ Uranium–Plutonium Mixed Fuel Fission Products on a Living Organism

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ABSTRACT

The article describes the results of experiments conducted on pigs to determine the effect of plutonium, which is the most radiotoxic and highly active element in the range of mixed fuel $(U_{0.8}Pu_{0.2})O_2$ fission products, on living organisms. The results will allow empirical prediction of the emergency plutonium radiation dose for various organs and tissues of humans in case of an accident in a reactor running on mixed fuel $(U_{0.8}Pu_{0.2})O_2$.

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1. Introduction

The main objective of this study is the prediction and assessment of radiation risks to humans from a nuclear reactor accident with mixed fuel $(U_{0.8}Pu_{0.2})O_2$ consisting of uranium (80%) and plutonium (20%). A subsequent assessment of the safety of rescue operations in the epicenter of a radiation accident at a reactor using mixed fuel $(U_{0.8}Pu_{0.2})O_2$ shall be conducted taking into account a dose of single external exposure.

Using the irradiation apparatus, shown in Fig. 1, it was found that the radioisotope composition of emergency emissions from a mixed fuel reactor core, constituting a radiation dose to humans, is rich in a stable compound of PuO_2, as well as a large amount of pure plutonium ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu,

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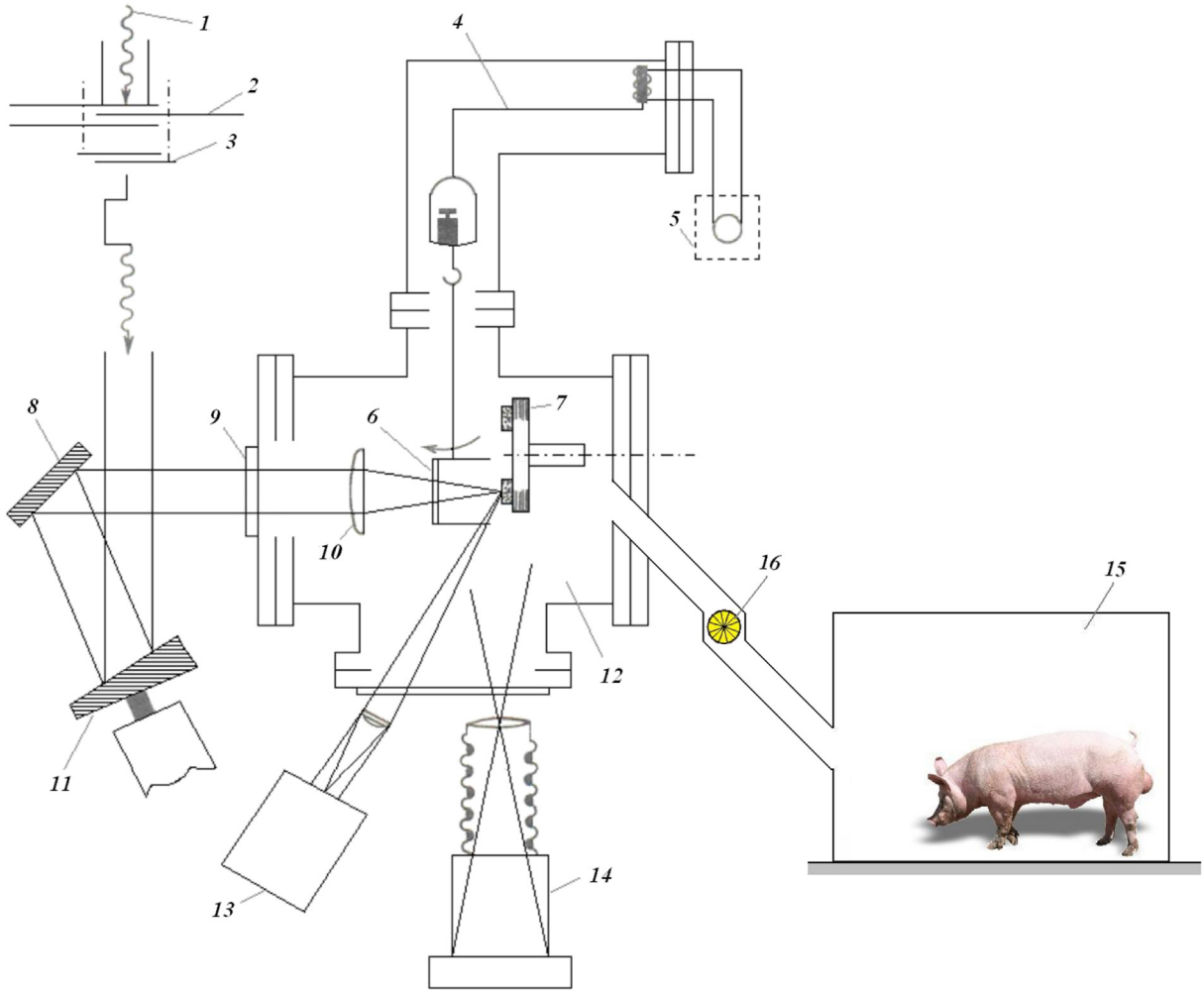

Fig. 1 – Schematic diagram of the installation for laser-induced heating of the samples of nuclear fuel to a temperature > 2,000°C. 1, CO_2 gas laser beam; 2, plasma opening switch (POS); 3, shutter; 4, vacuum balance; 5, compensatory circuit; 6, ballistic collector with an aperture for a beam; 7, turntable for targets; 8, stationary mirror; 9, aperture for a beam; 10, lens; 11, movable mirror; 12, vacuum chamber; 13, fast-acting micropyrometer for measuring the temperature in the focal spot and the evaporation period; 14, gas-jet observation camera; 15, plexiglass chamber for experimental animals; and 16, fan providing the influx of fumes from Chamber 12 into Chamber 15.

and ^{242}Pu, which are long-lived and the most radiotoxic of the expected emission components [1]. The irradiation apparatus is mounted at K.I. Satpayev Kazakh National Technical University (Almaty, Republic of Kazakhstan).

When calculating radiation doses as a result of leakage of fission products from a mixed fuel reactor core, it is necessary to take into account complex, diverse, and interrelated factors related to plutonium behavior. For calculations, it is necessary to have data or make appropriate assumptions on exposure conditions, including the composition of radioactive products, particle sizes, concentration of activity, and exposure time.

2. Materials and methods

During the experiments, the pigs were first subjected to artificial radiation treatment with plutonium; this was followed by an in-depth investigation of the metabolism of radioactive substances in the living body. The pigs being tested were slaughtered, and spectrometric analysis of various exposed organs and tissues were carried out for the subsequent evaluation of internal exposure of humans to products of salvo emissions from a mixed nuclear fuel reactor core. Spectrometric analysis of biological samples was conducted using the Progress-2000 measurement system, developed by Doza Research and Production Enterprise (Moscow, Russian Federation) and being a property of the Institute of Nuclear Physics of the National Nuclear Center of the Republic of Kazakhstan (Almaty, Republic of Kazakhstan).

To assess the degree of plutonium retention in human tissues, organs, and their systems, experiments were conducted on pigs because the human body has more similarities with a pig body compared with other land mammals. According to the Institute of Molecular Biology of the Academy of Sciences of the Russian Federation, announced at the VI International

Seminar on Individual Dosimetric Control (IWIMIR-2010) [2], humans and pigs have almost the same content of hemoglobin and proteins in blood, as well as the size of red blood cells and blood groups. Pigs, like humans, are omnivorous; they have similar digestion processes. Human skin is similar to that of a pig's—it can sunburn. Moreover, the two share many common features, such as the structure of teeth, eyes, liver, and kidneys. A pig heart weighs 320 g, whereas a human heart weighs 300 g. Pig lungs weigh 800 g whereas human lungs weigh 790 g, a pig kidney weighs 260 g whereas a human kidney weighs 280 g, and a pig liver weighs 1,600 g whereas a human liver weighs 1,800 g. Diseases of newborn piglets are similar to diseases of infants, and the somatotropic hormone molecular structure of pigs and humans is 70% alike.

In total, 32 pigs were involved in the experiment. Three major paths of plutonium penetration in pigs were studied in the experiments: inhalation (with inhaled air), oral (with food), and contact (administration of subcutaneous injection).

For inhalation of plutonium by the pig body, experimental animals were placed in airtight plexiglass chamber 15, where plutonium was pumped through plenum ventilation 16 formed in vacuum chamber 12 of the irradiation apparatus as shown in Fig. 1. The continuous plutonium inhalation period was on average 2 hours depending on the respiration rate of each experimental animal. The volume of air inhaled by pigs in a single breath ranged from 0.4×10^{-3} m^3 to 0.5×10^{-3} m^3, and the respiration rate ranged from 15 breath/min to 20 breath/min; therefore, within 2 hours about 1 m^3 of air containing plutonium penetrated into the pig's body, in order to ensure appropriate samples for the spectroscopic analysis.

In the course of the experiments on laser-induced heating of the samples of mixed fuel $(U_{0.8}Pu_{0.2})O_2$ with a total mass of 1 kg to a temperature above 2,000°C, it was found that the total volumetric activity of the plutonium contained in the resulting evaporations was 54 PBq/m^3 (Table 1). Thus, 54 PBq of plutonium was inhaled by the pigs within 2 hours.

Ingestion of plutonium in the pig's body was provided by feeding the experimental animals with grain forage previously placed in vacuum chamber 12 of the irradiation apparatus over the sample of nuclear fuel $(U_{0.8}Pu_{0.2})O_2$ and irradiated by fumes containing isotopes of plutonium—^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu [1]. The specific activity of plutonium-239 in the irradiated feed was 12 PBq/kg (12×10^{15} Bq/kg); the levels of content of other plutonium isotopes did not exceed the level of spectrometer sensitivity. The feed dosage of each pig was 1 kg, or 12 PBq of plutonium-239 in the body of each experimental animal.

Contamination from the excreta of pigs was also inevitable because samples of urine and feces were required for the studies. It was found that about 30% of plutonium is excreted with feces during 12 hours after intramuscular injection. The major part of the deposited plutonium is excreted with the urine and less is excreted with the feces. Urine was collected via silicone containers attached to the torso and fixed porcine peritoneum below the urethra. Feces were collected throughout the cage, wherever the pigs excreted their wastes. The collection sites were deactivated immediately after feces sampling by means of the Petrov contact, Trilon B, and other deactivating agents. Sample preparation was carried out by urine evaporation, washing the precipitation with a cotton swab dipped in a solution of nitric acid, and subsequent ashing of the swab. The ash was subjected to spectrometric analysis. The feces were dried in an oven at 200°C to a powder state, and then were sent for spectrometric analysis.

Because the skin of the experimental pigs had no damage, the artificial infliction of which is prohibited by the European Convention for the Protection of Vertebrate Animals Used for Experimental and other Scientific Purposes (ETS N 123, Strasbourg, March 18, 1986), contact administration of plutonium in pigs was conducted via an intramuscular injection of 20 mL of physical solution with plutonium-239 concentration equal to 1 g/mL.

Unfortunately, the skin of pigs placed in the chamber had no protection from surface contamination. This introduced a certain error in the experimental purity, as the contamination of the skin surface inevitably led to the ingestion of radionuclides. However, neglecting the protection of the skin could be partly justified by two factors: (1) the purpose of the research was to assess the internal dose of a living organism, which is formed by oral intake of radionuclides *inter alia*; and (2) because the subject of the study was the dose formed by plutonium, which is an alpha emitter with the particles' mileage in biological tissue about 31–56 μm, the protection of the skin (thickness of pigs' skin is 1–3 mm) from contact exposure was not necessary.

After the pigs' exposure in the chamber, swabs were taken from their skin surface. The levels of skin contamination exceeded neither 2 alpha particles/(cm^2 × min) nor 200 beta

Table 1 – Volumetric activities of plutonium radionuclides contained in $(U_{0.8}Pu_{0.2})O_2$ fuel fumes at 2,000°C.

Nuclide	Half-life (hr)	PVA (Bq/m^3)	Measured volumetric activity of a nuclide, 10^{15} Bq/m^3	Nuclide activity, % to the total activity of evaporations
^{238}Pu	7.68×10^5	5.30×10^{-1}	2	0.0003
^{239}Pu	2.11×10^8	5.30×10^{-1}	24	0.002
^{240}Pu	5.73×10^7	5.30×10^{-1}	8	0.0009
^{241}Pu	1.26×10^5	5.00×10^{-1}	13	0.001
^{242}Pu	3.29×10^9	5.70×10^{-1}	7	0.0007
Total volumetric activity of plutonium			54×10^{15} Bq/m^3	

Note. From "Spectrometry analysis of fumes of samples of mixed nuclear fuel (U0.8Pu0.2)O2 heated up to the temperature 2,000°C," by M.T. Zharaspayev and D.S. Kim, 2012, Instruments and Technique of Experiment, pp. 1–5. Academy of Science of the Russian Federation, Publishing House "Nauka," Moscow. Copyright 2012, Copyright Holder – Pleiades Publishing, Ltd. Reprinted with permission. Column 3 from "99 SP 2.6.L.758-99, Radiation standards, Official issue, Intr. 1999-01-01," by Agency on Health Protection Affairs of the RK, Almaty, 1999. Copyright 1999, Name of Copyright Holder – Ministry of Health of the RK. Reprinted with permission.
hr, hour; Pu, plutonium; PVA, permissible volumetric activity.

particles/(cm^2 × min), and so were in fairly good agreement with the requirements on radiation protection [3].

Of course, the intake of radionuclides through the skin makes a minimum contribution to the internal dose of irradiation, but if the skin is damaged, then a large amount of radioactive particles could penetrate the body through a wound. Injection of solution (HNO_3Pu) simulated the contact intake of plutonium through damaged skin directly into the bloodstream.

Because calcined silica is difficult to dissolve in acids, the dissolved plutonium was obtained by electrolytic dissolution using divalent silver as an intermediate oxidation product.

PuO_2 was injected into HNO_3 (ranging from 4N to 6N), containing $AgNO_3$. The solution was passed through the anode compartment of an electrolyzer. The generated ^{11}Ag oxidizes the plutonium dioxide PuO_2, which is then dissolved in nitric acid. Reduction reactions occurred near the cathode (especially restoration of NO^{3-}). Consumption of nitric acid must be compensated for by continuously adding into the cathode compartment (where the normality 8N and more is supported to avoid hydrogen release).

3. Results and discussion

The data in Table 1 show that among the plutonium isotopes, the dominant contribution to the total activity measured in vapors above uranium–plutonium oxide $(U_{0.8}Pu_{0.2})O_2$ is made by plutonium-239 [1]. It is logical that this radioisotope will be deposited *in vivo* the most, and its ingestion will subsequently require integrated medical and rehabilitation measures.

In order to develop a course of treatment for excretion of radiotoxic plutonium from the body, it is necessary to calculate the radiation doses for the main locations of isotope accumulation.

As a result of the experiments on pigs, it was found that most of the plutonium entered the body via inhalation and is deposited mainly in the lungs, bones, liver, and in the reproductive organs.

At present, the internal exposure dose of internal organs and bones from the particles of any radioactive substance that penetrated into the body is calculated based on the assumption that the radiated energy is evenly distributed throughout the organ tissue [4]. The average value of the dose for the whole organ is taken as the resulting value.

Plutonium dioxide emits alpha particles with an energy of 5.14 MeV and a free path through tissue of < 40 μm, recoil atoms with an average energy of ~0.084 MeV, and low-energy X-ray and gamma ray radiation characterized by an energy of < 0.01 MeV for decay. It is considered that any organ absorbs all alpha particles and recoil atoms, and a portion of the X-ray and gamma radiation depending on organ size [5].

Effective radiation energy ($E_{eff.}$) is calculated for each particular radioisotope in a single organ. In the case of uniform deposition of radioactive substances in each of several organs, the absorbed dose can be calculated. For convenience of calculations, the obtained values of effective energy are shown in Table 2 [6].

However, radiation loads from the particles of a radioactive substance that penetrated into the body should be evaluated based on the equivalent (not absorbed) dose, because, first, various types of radiation have different characteristics. Second, in most cases radioactive substances are unevenly distributed in the tissue.

For radiation protection, the equivalent dose (D_{eqv}, μSv) is defined as the product of the absorbed dose (D_{abs}, μGy), radiation weighting factor (WR), and coefficient of dose distribution (CD) [7]:

$$D_{eqv} = D_{abs} \times WR \times CD \quad (1)$$

The equivalent dose of 1 Bq · hr is determined using the following equation:

$$\frac{D_{eqv}}{Bq \cdot hr} = 51.2\frac{E_{eff}}{m}, \quad (2)$$

where E_{eff} is the effective radiation energy (MeV) and m is the mass of the organ (g).

The coefficients given in Table 2 were used to determine the effective energies of the other components of the mixed nuclear fuel. The effective energies for each of the fission products of $(U_{0.8}Pu_{0.2})O_2$ [8] in the lungs, bone tissue, liver, and ovaries are given in Table 3.

In laboratory conditions of exposure, it was found that some fraction of inhaled plutonium (f_a) is deposited directly into the lungs, wherein 40% of the initial amount of deposited plutonium is eliminated with an effective half-life of 1 day (24 hours) and the remaining 60% with an effective half-life of 500 days (12,000 hours).

Only small amounts of plutonium get in the blood by inhalation. The low solubility of plutonium in the fluids covering the respiratory tract leads to its deposition and rapid release from the nasopharyngeal and tracheobronchial regions. Nevertheless, prolonged retention of particles in the lung region leads to absorption of ~5% of PuO_2 in the blood; ~15% of the deposits in the pulmonary region transferred to

Table 2 – Values of the effective energy of different types of radiation emitted by ^{239}Pu.

Radiation type	Energy, E (MeV/decay)	Weighting factor, WR	$E_{eff.}$ (MeV)		
			Bone tissue	Gastroenteric tract	Other organs
Alpha radiation	5.14	10	257	0.51	51.4
Knock-on atoms	0.084	20	8.4	0.01	1.68
X-ray and gamma radiation	0.0085	1	0.04		
Total			270	0.52	53.0

Note. From "The chemical thermodynamics of nuclear materials: IX. The high temperature heat capacity of plutonium-3.2 at. % gallium alloy," by R.O. Adams and F.L. Oetting, 1983, *J. Nucl. Mater.*, 118, p. 269–274. Copyright 1983, Copyright Holder Elsevier B.V. Reprinted with permission. E_{eff}, effective radiation energy; Pu, plutonium.

Table 3 – Effective energies of various components of mixed fuel $(U_{0.8}Pu_{0.2})O_2$ [8] contained in the lungs, bone tissue, liver, and ovaries.

Radioisotope	Radioisotope activity portion (Bq/Bq of isotope mix)	$E_{eff.}$, MeV			
		Lungs	Bone tissue	Liver	Ovaries
^{238}U	1.9×10^{-2}	43	220	43	0.43
^{237}Np	1.36×10^{-4}	49	250	49	0.49
^{238}Pu	2.83×10^{-3}	53	270	53	0.53
^{239}Pu	2.46×10^{-2}	57	284	57	0.57
^{240}Pu	8.58×10^{-3}	53	270	53	0.53
^{241}Pu	1.3×10^{-2}	0.53	14	1.0	0.02
^{242}Pu	6.78×10^{-3}	51	250	51	0.51
^{241}Am	5.89×10^{-4}	57	283	57	0.57
^{243}Am	3.31×10^{-4}	54	272	54	0.54
^{242}Cm	1.84×10^{-3}	64	400	78	0.78
^{244}Cm	5.89×10^{-4}	60	300	60	0.60

Am, americium; Cm, curium; E_{eff}, effective radiation energy; Np, neptunium; Pu, plutonium; U, uranium.

pulmonary lymph nodes eventually reach the circulatory system. Thus, ~20% of the particles deposited in the pulmonary region after a while will be transferred to other organs via the blood or circulating lymph.

Therefore, intake of 1 Bq of plutonium leads to irradiation by dose I [9,10]:

$$I = f_a\left(\frac{40\% \times 24 \text{ hours}}{0.693} + \frac{60\% \times 12,000 \text{ hours}}{0.693}\right) = 10,404 f_a, \text{ Bq/hr}, \quad (3)$$

where f_a is the activity of plutonium that entered the pig's body with inhaled air in Bq.

The values of the internal exposure dose of a human being largely depend on the mass of their organs and their respiration rate, which in turn are subject to age-related changes. The radiation dose from plutonium to the internal organs, accumulated by the time the patient will be 70 years old, depending on the age, when plutonium-239 penetrated into the body, can be calculated using the following equation [9,10]:

$$D_{1,2} = K\left[\frac{t_2 - t_1}{\ln m_2 - \ln m_1}\left(\frac{1}{m_1} - \frac{1}{m_2}\right)\right] \text{ μSv/Bq}, \quad (4)$$

where $D_{1,2}$ is the dose of multiple human exposures, and K is the correction factor depending on the organ and tissue: $K = 47.36 \times 10^5$ for lungs [10], $K = 5.30 \times 10^5$ for bone tissue [9], and $K = 1.39 \times 10^5$ for liver [9].

Calculation results are given in Table 4.

Besides ^{239}Pu, there are other plutonium isotopes in the mixed fuel of a nuclear reactor with relative amounts depending on the nature of the fuel rods used. If we consider the effects of plutonium on a human apart from all other radioisotopes, the radiation doses can be calculated on the basis of the data given in Table 5.

If the share of inhaled plutonium (f_a) is not determined, the radiation dose from plutonium deposited in the lungs can be determined using the following equation [11]:

$$I = \frac{1}{0.693}\left\{0.4\frac{24T_{1/2}}{T_{1/2}+24}\left[1 - e^{-0.693t}\left(\frac{T_{1/2}+24}{24 \times T_{1/2}}\right)\right] + 0.6\frac{12,000T_{1/2}}{T_{1/2}+12,000}\left[1 - e^{-0.693t}\left(\frac{T_{1/2}+12,000}{12,000T_{1/2}}\right)\right]\right\} \quad (5)$$

where $t = 50 \times 365 \times 24 = 438,000$ hours and $T_{1/2}$ is the half-life in hours.

Thus, plutonium radiation exposure by particles in the lungs would be equal to 10,389.46 Bq • hr per Bq of accumulated plutonium:

$$I = 1.443 \times [9.5999 + 7,190.3052] = 10,389.46\frac{\text{Bq·hr}}{\text{Bq}_{Pu}} \quad (6)$$

However, only 0.025 of the total activity of the plutonium isotopic mixture falls on ^{239}Pu, so the contribution of ^{239}Pu to the total dose will be 259.74 Bq (^{239}Pu) • hr per Bq of the mix of isotopes accumulated in the lungs [7]:

$$I = 0.025 \times 10,389.46 = 259.74\frac{\text{Bq}_{^{239}Pu}\cdot\text{hr}}{\text{Bq}_{mix}}, \quad (7)$$

In this case, the daily dose from each Bq of ^{239}Pu deposited in the lungs will be 3.82 μSv [7]:

$$\frac{CD}{\text{Bq}_{^{239}Pu}} = 51.2 \times \frac{E_{eff}}{m} = 51.2\frac{57}{764} = 3.82\frac{\mu\text{Sv}}{\text{Bq·hr}}, \quad (8)$$

and the accumulated dose (AD) due to ^{239}Pu will be almost 1 mSv per Bq of isotopic mixture deposited in the lungs [7]:

$$AD_{^{239}Pu} = 259.74 \times 3.82 = 992.19\frac{\mu\text{Sv}}{\text{Bq}} = 0.992\frac{\text{mSv}}{\text{Bq}}. \quad (9)$$

Table 6 shows the predicted values of radiation doses to the lungs, bone tissue, liver, and ovaries of people exposed to radiation at 20 years of age, depending on the activity of the isotopic mixture deposited in the lungs, at the end of 50 years.

Data on the total radiation dose to the lungs, skeleton, liver, and ovaries from an isotopic mixture of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu accumulated in the lungs are given in Table 7.

The values shown in Tables 6 and 7 were obtained via Eqs. (6–9), i.e., these are the theoretical values, which could not be approximated to integers, because one is unlikely to take into account the difference in the assessment of radiation exposure from the different plutonium isotopes in various organs and tissues. The measurement results of volumetric activity of plutonium isotopes are represented by integers in Table 1, but only if the coefficients of measurements error were < 40%.

Table 4 – Radiation doses of human lungs, bone tissue, and liver expected by age 70, depending on time of ^{239}Pu penetration in the body.

Age when activity has penetrated into the body (yr)	Proposed mass (g)			Average respiration rate (m^3/hr)	Radiation exposure (μSv/Bq of accumulated activity)		
	Lungs	Bones	Liver		In lungs	In bones	In liver
Newborn	66	500	100	0.033	0.03	0.09	0.15
1	170	2,000	250	0.16	0.02	0.09	0.13
10	450	5,900	844	0.62	0.009	0.07	0.09
20 (female)	764	6,800	1,450	0.88	0.006	0.06	0.07
20 (male)	955	10,000	1,810	0.95	0.005	0.04	0.06

hr, hour; Pu, plutonium; yr, year.

Table 5 – Expected composition of plutonium isotopic mixture in the fuel.

Radioisotopes	Share of activity[a] (Bq/Bq of isotopes mix)	Half-life (hr)	Dose per unit of activity, deposited in the bone tissue, liver, & ovaries (μSv/Bq of accumulated activity)
^{238}Pu	0.0028	7.9×10^5	0.06
^{239}Pu	0.025	2.1×10^8	0.07
^{240}Pu	0.0085	5.8×10^7	0.07
^{241}Pu	0.013	1.1×10^5	0.02
^{242}Pu	0.0068	3.4×10^9	0.07
Total			0.29

[a] Specific activity is 17.113×10^{12} Bq/kg of plutonium isotopic mixture [12].
Pu, plutonium.

The systemic form of plutonium is distributed mainly in the bone and in the liver. After the intramuscular injection of plutonium, the authors found that ~60% of the absorbed plutonium is deposited in bones, ~4% in the liver, and 30% is gradually excreted. The results, which were obtained from the experiments, have shown a much higher potential risk of deposition of plutonium in the liver. Our investigations revealed that only 20% of the plutonium that had been deposited in a pig's body was excreted out of the liver in 4 years. The time of deposition of plutonium in the liver does not significantly differ from the time of its deposition in bones.

The reference value of the specific activity of a plutonium isotopic mixture in biological tissue is equal to 17.113 TBq/kg (17.113×10^{12} Bq/kg) [12], and using Eq. (10) we can calculate the total radiation dose to the lungs, skeleton, liver, and ovaries of a human in relation to the mass. Eq. (10) connects the mass of any radionuclide with its activity value [13].

$$m = b \times Z \times T_{1/2} \times A, \tag{10}$$

where m is the mass of the radionuclide in g; Z is the atomic mass of the radionuclide; A is the activity of the radionuclide in Bq; $T_{1/2}$ is the half-life of the radionuclide; and b is the factor depending on the measurement unit of the half-life: $b = 8.40 \times 10^{-24}$ if $T_{1/2}$ is measured in seconds, $b = 1.44 \times 10^{-22}$ if $T_{1/2}$ is measured in minutes, $b = 8.62 \times 10^{-21}$ if $T_{1/2}$ is measured in hours, $b = 2.07 \times 10^{-19}$ if $T_{1/2}$ is measured in days, and $b = 7.56 \times 10^{-17}$ if $T_{1/2}$ is measured in years.

According to the results of the preceding equation, the activities of plutonium isotopes correspond to the following

Table 7 – Total radiation doses to lungs, bone tissue, liver, and ovaries depending on activity of isotopic mixture accumulated in the lungs (units—mSv/Bq of isotopes mix accumulated in lungs).

	Mass (g)	Deposited dose of plutonium penetrated into the body by inhalation (mSv/Bq of isotopic mixture deposited in the lungs)	Total radiation dose (mSv/Bq of isotopic mixture deposited in the lungs)
Lungs	764	–	1.65
Bone tissue	6,800	0.39	0.78
Liver	1,450	0.59	0.87
Ovaries	8	0.00093	1.59

Table 6 – Radiation doses to lungs, bone tissue, liver, and ovaries depending on activity of isotopic mixture accumulated in the lungs (unit of measurement—mSv/Bq of isotopic mixture deposited in the lungs).

Radioisotope	Radiation dose to the lungs	Radiation dose to the bone tissue	Radiation dose to the liver	Radiation dose to the ovaries
^{238}Pu	0.103	0.059	0.054	0.098
^{239}Pu	0.992	0.555	0.522	0.947
^{240}Pu	0.313	0.018	0.165	0.299
^{241}Pu	0.005	0.014	0.005	0.017
^{242}Pu	0.241	0.132	0.127	0.230
Total	1.65	0.78	0.87	1.59

Pu, plutonium.

values of its mass: 1 Bq of $^{238}Pu \equiv 1.58 \times 10^{-12}$ g; 1 Bq of $^{239}Pu \equiv 4.35 \times 10^{-10}$ g; 1 Bq of $^{240}Pu \equiv 1.19 \times 10^{-10}$ g; 1 Bq of $^{241}Pu \equiv 2.62 \times 10^{-13}$ g; 1 Bq of $^{242}Pu \equiv 6.88 \times 10^{-19}$ g; and total mass $= 5.56 \times 10^{-10}$ g.

The results of calculations of the total radiation dose to lungs, bony skeleton, liver, and ovaries of a human in relation to the mass of plutonium isotopic mixture accumulated in the lungs are given in Table 8.

The data in Tables 6–8 show that plutonium absorption from the lungs causes the deposition of 31% of the amount that penetrated the body in the lungs, 30% in the ovaries, 17% in the liver, 15% in the skeleton, and 7% in other tissues.

Spectrometric analysis of biological samples of pigs slaughtered a day after feeding with grain contaminated with plutonium showed that absorption of a small amount of PuO_2 from the gastrointestinal tract occurs as a result of partial dissolution. The upper limit of the intestinal absorption of plutonium was 0.007% of the activity entering the pig's body orally, and the degree of digestion was not more than 0.003%. Such a low percentage of plutonium absorption can be explained by the fact that the intestinal mucosa is an effective barrier for plutonium absorption during exposure. Plutonium absorbed from the gastrointestinal tract comes into the blood in ionic or diffusing complex form and as a result is deposited in the bones.

When assessing the internal exposure of pigs from contact with plutonium, the most vulnerable external organs, the eyes, were considered. The rate of deposition of plutonium particles on the skin surface under normal conditions is about 5×10^{-3} m/s, and the area of the pig eye is about 4×10^{-4} m^2. Thus, the total plutonium absorption by the pig eyes should be taken into account twice because pigs have two eyes (Bq · s/m^2). However, it is known that the greatest danger for living organisms is contact introduction of radioactive substances through damaged skin. To simulate the introduction of plutonium through epidermal damage, intramuscular injection of a concentrated solution containing ^{239}Pu was administered to pigs.

The results of the spectrometric analysis of biological samples from pigs slaughtered 12 hours after the intramuscular injection of plutonium showed that about 60% of absorbed plutonium was deposited in the bones, 4% in the liver, and 30% is excreted.

To quantify the excretion of plutonium from the living body, 12 experimental animals exposed to plutonium via inhaled air, in the feed, and in the injected solution, were slaughtered 4 years after the initial experiments. Prior to the experiments, faster excretion of plutonium was predicted from the liver than from other organs and bone tissue, but the results obtained after the experiments showed a much higher plutonium potential deposition in the liver. It turned out that within 4 years only 20% of the deposited plutonium was excreted from the liver. The time of plutonium retention in the liver does not differ significantly from the time of retention in the bone tissue. Thus, the initial conservative assumption that there is no substantial excretion of plutonium from the liver is apparently true.

Plutonium concentration in the ovaries is approximately 20 times higher than that in skeletal muscles, and approximately six times higher than that in the heart. This was true for both monomer and polymer plutonium even 4 years after the initial exposure.

The experiments proved that the rate of plutonium excretion from the main locations of deposits in a living organism is very low, so when assessing internal exposure doses, biological excretion may be neglected; only therapeutic plutonium excretion acceleration is of practical interest.

Table 8 – Total radiation doses to lungs, bone tissue, liver, and ovaries depending on activity of isotopic mixture accumulated in the lungs (units—mSv/kg of isotopes mix accumulated in the lungs).

	Mass (g)	Total radiation dose (mSv/kg of isotopic mixture deposited in the lungs)
Lungs	764	15.70
Bone tissue	6,800	7.42
Liver	1,450	8.28
Ovaries	8	15.13

3.1. *Biokinetic data and observation of biological effects*

Many modern problems of determining the expected level of exposure in a living organism after an accident in a mixed fuel reactor, as well as the exact influence of ionizing radiation on any kind of biological tissue, remain unexplored.

Ionizing radiation effects on the living organism cause molecular dissociation and formation of new compounds under the influence of free radicals, and lead to disturbances in the cellular structure of biological tissue. The novel compounds encountered in living cells by free radicals cause contravention of cell division kinetics, gene damage, or cell death. If persistent changes occur in the genetic apparatus of germ cells, the result could be a mutation in the offspring of an irradiated specimen.

Changes in the cell structure lead to disruption of metabolic processes in the body or to the early physiological effects. This would cause dysfunction of tissues and organs, resulting in the death of the entire organism.

In simple substances, molecules consist of a single kind of atom, and the process of ionization is accompanied by the process of recombination. The ionized atom attaches itself to one of the free electrons, which are always present in the medium, and the neutral atom is formed again. An excited atom returns to the normal state by the transition of an electron from the outer shells to the vacant place of the nearest shell to the nuclei of the atom. The ionization and excitation of atoms of simple substances do not lead to any changes in the physical and chemical nature of the irradiated medium.

The situation is different when ionizing radiation influences complex organic substances, the molecules of which consist of a large number of particular atoms. The molecules remain in the excited state for 10^{-13}–10^{-14} seconds [14]. During this time, the excitation energy is converted into oscillatory energy and focuses on one of the chemical bonds, and this leads to the collapse of the molecule and the avulsion of any fragment from it. The consequence of ionization is an abrupt change in the electromagnetic field of the molecule, which

leads to a break of 10–15 chemical bonds, i.e., during the processes of ionization and excitation of complex molecules, disintegration occurs as a result of breaking chemical bonds. In organic molecules the carbon bonds are broken, and new molecules—often alien to the body structure—are formed.

The indirect influence of ionizing radiation plays an important role in the formation of radiation-induced effects in biological tissue. Indirect influence implies the radiation-chemical changes caused by water radiolysis products, forming free radicals H^- and very active OH^-:

$$\begin{aligned} H_2O^+ &\rightarrow H^+ + OH^0 \\ H_2O^- &\rightarrow H^0 + OH^- \end{aligned} \quad (11)$$

The indirect influence of ionizing radiation causes a shift in the acid–base balance and changes in redox processes that lead to metabolic disorders in the body.

Biological tissue consists of 60–70% water. Free radicals react chemically with the molecules of protein, enzymes, and other structural components of biological tissue, leading to changes of the biological processes in the body. As a result, metabolism is violated, the activity of enzyme systems is inhibited, the growth of tissues slows down and stops, new chemical compounds (toxins) arise, and all of this leads to disruption of vital functions or systems of the whole body.

The most dangerous injuries damage mitosis and chromosomal apparatus. The number of damaged cells in the irradiated population depends on the absorbed dose of radiation, blocking of physiological processes of regeneration, and body vitality. The cellular changes are likely to disrupt the DNA structures, inhibit hematopoietic function, and suppress spermatogenesis.

Under the influence of the energy of radioactive particles and electromagnetic waves, the formation of a wound surface and rupture of chromosomes can occur. In most cases, the cells are killed at the same time, but in rare cases the cells with damaged chromosomes divide and give rise to a new tissue (tumor). As a result of cell death, tissues cannot cope with their functional tasks, and decompensation of its functions appears [15].

To assess the retention of plutonium in various tissues, vital organs, and systems, the three main routes of plutonium matriculation into the living organism have been investigated: respiratory tract, digestive tract, and contact route. Schematically, the routes of plutonium matriculation into the human body can be presented as shown in Fig. 2 [16].

The absorption ways, significant for the soluble compounds only, are shown in dashed lines.

Internal irradiation caused by the release of plutonium in the environment depends on the physical, chemical, and biological properties of the extracted material; the local environment; the organism of a human; and the properties of the release. Distribution and retention of plutonium in living organisms have been studied on pigs. The results are not fully consistent with each other, and one can formulate only a few general conclusions concerning the early distribution of matriculated plutonium compounds. In all cases, the primary deposition areas are bone, liver, and gonads. Conditions favoring the absorption of plutonium in a monomeric state lead to a higher rate of bone deposition, whereas polymeric forms tend to be delayed in the liver. Although small amounts of plutonium are deposited in other tissues, the concentration of these deposits is not considered significant.

Plutonium is transferred by blood to vital organs and systems. In turn, plutonium reaches the bloodstream in three different forms [17,18]: (1) in easily diffused form, which could be either ionic or complex; (2) in combination with relatively large proteins that do not diffuse from the circulatory system; and (3) in the form of larger colloidal aggregates that can enter the bloodstream or directly form in the blood after absorption of some ionic species.

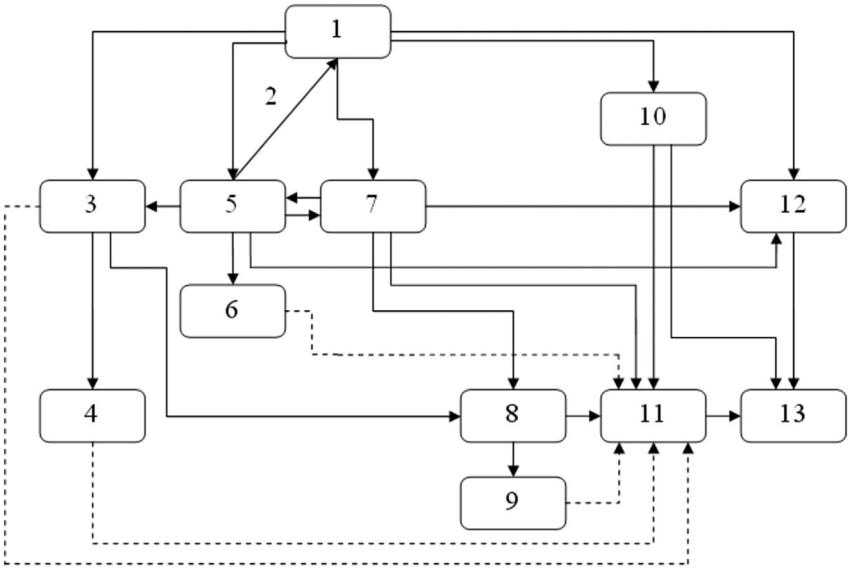

Fig. 2 – Origins of human exposure to radioactive materials. 1, scattering material; 2, suspension; 3, surface water; 4, fish and seafood; 5, soil; 6, groundwater; 7, vegetation; 8, meat of animals; 9, animal products; 10, inhalation; 11, ingestion; 12, skin, eyes; and 13, dose received by human. *Note.* From *The Mechanics of Aerosols*, by N.A. Fuchs, 1964, Pergamon, Oxford, Chap. 11. Copyright 1964, Copyright Holder-Elsevier B.V. Reprinted with permission.

To obtain information about the forms of plutonium distribution in the tissues of a living organism, experiments were conducted on pigs, and the survey results obtained during autopsy of cadavers that have been exposed or not exposed to plutonium were also taken into account.

Plutonium, absorbed from the gastrointestinal tract of pigs, penetrated the body as a result of intramuscular injection, and absorbed through the intact skin, entered the bloodstream in an ionic or complex diffusing form, and is deposited in bones. In studies conducted during autopsies and some experiments on PuO_2 respiration using pigs, it was observed that plutonium is deposited mainly in the liver. It is assumed that plutonium can be absorbed from PuO_2 deposits in the lungs in the form of Pu^{4+} or in the form of macroparticles, or in both forms at once.

Deposition and retention of plutonium in bones deserve special attention. Research on specific concentrations of plutonium in various parts of the pig skeleton did not provide completely consistent results. However, the highest concentrations are found in the spine, and the lowest in the flat bones of the skull. Studies have shown that the deposition of plutonium in the skeleton is not dependent on an individuals' age.

The total amount of plutonium in the skeleton of experimental animals had been measured repeatedly. In the result of our study, which was conducted on seven pigs over a period of 4 years, we determined that the half-life of plutonium from the bone ribs is 9 years, and that from the femoral diaphysis is 11 years. In the results of a study that was carried out on 25 pigs, we came to the conclusion that about 15% of plutonium is taken out of the skeleton within 4 years, corresponding to a half-life of ~17 years.

Although direct measurements of the dynamics of plutonium deposition in the human skeleton have never been conducted, we can probably evaluate the half-life of plutonium extirpation proportionally to the general release rates of plutonium. It is believed that ~20% of the absorbed plutonium is lost from the body during the first 50 years because of natural excretion, i.e., the half-life is > 150 years. Therefore, to calculate the internal dose in bones, one can suggest that deposition in the skeleton would be kept permanently.

After administering an intramuscular injection of plutonium, the authors found that ~60% of absorbed plutonium is deposited in bones, ~4% in the liver, and 30% is gradually excreted. Such a distribution could be considered as approximately correct when plutonium enters the human body through wounds or through the gastrointestinal tract.

Besides the intraosseous retention of plutonium, its deposition was expected in the liver but only in very small amounts. During the early estimates of plutonium distribution in the body, it was assumed that 90% of the total plutonium amount would be deposited in the skeleton and only 7% in the liver. The distribution of plutonium in the body between the liver and the bone tissue depends on the chemical and physical state of the plutonium in the blood. However, because of the difference in the total weight of these two tissues, the plutonium concentration in the liver can be considered roughly the same or substantially higher than the concentration in the bone. Consequently, liver and bone are the main areas of plutonium deposits.

Initially, plutonium excretion from the liver was predicted to be more rapid than that from bone tissue [19,20]. However, results obtained after the experiments have shown a much higher potential risk of deposition of plutonium in the liver. Our investigations revealed that only 20% of the plutonium that had been deposited in a pig's body was excreted out of the liver in 4 years. The time of deposition of plutonium in the liver does not significantly differ from the time of its deposition in bones. Thus, the initial conservative assumption—that substantial excretion of plutonium out of the liver does not occur—seems true.

Argonne Cancer Hospital (University of Chicago, Chicago, IL, USA) experts argue that the exposure of ovaries could be considered as the decisive factor of the genetic dose assessment. The concentration of plutonium due to the radioactive fallout in the complex "gonadal" sample made up of samples that were taken from six corpses is second only to the concentration in the lung lymph nodes [9]. Spleen and kidney contained an average of 22.2 GBq/kg, whereas lung lymph nodes contained 181.8 GBq/kg, gonads contained 130 GBq/kg, and lungs contained 28.8 GBq/kg. In each case, death occurred after age 70 years.

At present, there are no data regarding the rate of plutonium excretion from the human gonads. However, from the results of experiments on pigs it was found that the half-life of plutonium from the ovaries is at least 450 days.

The ovaries of pigs accumulate 0.06% of a single intravenous injection of plutonium nitrate. In one pig, it was shown that 2×10^{-3}% of an intravenous injection of plutonium nitrate (IV) per 1 g of the ovaries was deposited. If we assume that the ovaries weigh about 40 g, the total amount of deposited plutonium would be about 0.2% of the administered dose. This result is confirmed with fairly good agreement by figures of the distribution function with the results of another study [21].

Plutonium nitrate (pH = 2–2.5) was injected intratracheally into little pigs. The amount of solution found in the ovaries of the two animals slaughtered on the 25th day after irradiation was ~0.02% of the total caught in the blood. It was also found that 0.068% of the absorbed plutonium was deposited per 1 g of the ovaries and the adrenal glands. Assuming that these organs weigh about 40 g, and if the amount deposited in the ovary is 75% of the total fat in the ovaries and adrenal glands, the results show that ~0.2% of the absorbed plutonium was deposited in the ovaries.

Based on the above research, it could be assumed that ~0.2% of the plutonium that enters the bloodstream is deposited in the gonads: ovaries or testes. The observed rate of plutonium excretion from ovaries or testes is low; even 40 weeks after exposure, there was no obvious reduction of plutonium in the pigs' ovaries. Deposits in the ovaries are mainly concentrated in the granular layer of some follicles.

Recent data, obtained during plutonium injection in pigs, have shown that the plutonium concentration in the ovaries is approximately 20 times higher than that in skeletal muscle, and approximately six times higher than that in the heart. This was true for both the monomer and polymer plutonium even 1.5 years after irradiation. In fact, the plutonium concentration in the ovary was comparable to the plutonium concentration in the liver when the monomeric form was administered, whereas injection of the polymer form increased the plutonium concentration in the liver above the plutonium concentration in the ovaries.

3.2. *Conclusion*

A long-term forecast of internal exposure by a mixture of plutonium isotopes is made taking into account a single inhalation of the particles in the body and their subsequent distribution to individual organs. The dose per unit activity of ^{239}Pu (Bq) deposited in the bone tissue, liver, and ovaries was 3 mSv. The entire range of plutonium isotopes formed a radiation dose equal to 4.89 mSv/Bq of deposited activity.

Doses of internal irradiation were predicted from plutonium, in vapors of 1 kg of mixed nuclear fuel $(U_{0.8}Pu_{0.2})O_2$, heated to 2,000°C. The dose from plutonium deposited in the lungs 50 years after the accidental exposure of a person would be 15.7 mSv for the lungs, 7.42 mSv for the bone tissue, 8.28 mSv for the liver, and 15.13 mSv for the ovaries.

Thus, human internal plutonium exposure doses contained in evaporations of mixed nuclear fuel $(U_{0.8}Pu_{0.2})O_2$ with a mass of 1 kg, heated to a temperature above 2,000ºC, would not exceed the permissible limits of radiation safety standards for workplace exposure [3]. However, the fuel element column of a nuclear power reactor core can exceed 50 tons; in case of accidental exposure to nuclear fuel fission products of such mass, radiation doses for the human body would increase by several thousand times.

Conflicts of interest

The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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