Atomic Energy, Vol. 78, No. 6, 1995

ESTIMATION OF THE EMISSION OF RADIOACTIVE SUBSTANCES DURING THE 1986 ACCIDENT IN THE FOURTH-POWER GENERATING UNIT AT THE CHERNOBYL NUCLEAR POWER PLANT (REVIEW OF PRIMARY DATA)

Yu. V. Sivintsev and A. A. Khrulev

UDC 621.039.586

Immediately after the first information about the accident at the Chernobyl nuclear power plant was received, three groups of experts were formed at the I. V. Kurchatov Institute of Atomic Energy. The purpose of these groups was to analyze the accident. Later they were given official status by a special decree of the President of the Academy of Sciences of the USSR. The group of which the authors of the present report as well as O. Ya. Shakh were members was charged with estimating the activity, composition, and dynamics of the emission of radioactive substances from the damaged power-generating unit on the basis of computational, experimental, and field data.

All estimates presented below were obtained in May–June 1986 [1–6]. The results of the analyses were reported immediately to the leadership of the Institute, the Presidium of the Soviet Academy of Sciences, a government commission, and the Politburo of the Central Committee [4, 5]. These results formed the basis of the corresponding section of the report presented by Soviet experts in July 1986 at the IAEA [7].

An objective estimate of the parameters of the emission of fission products and fuel from the damaged reactor can be made only if the following parameters are known:

activity of radionuclides accumulated in the core;

character and dynamics of accident development;

state of the fuel and reactor as a whole; and,

region (zone) of propagation and composition of the radioactive fallout.

The situation was especially difficult because there was very little initial information. Only computational data on the activity of the radionuclides accumulated in the RBMK reactor during normal operation [8], which differed substantially from the real activity, were available. The primary information received about the real radiation conditions and the composition of the samples was sporadic and random. This, together with the lack of practical experience in estimating the consequences of serious radiation accidents, made it impossible to formulate unequivocal conclusions and often resulted in divergent points of view. For this reason, the working documents on the assessment of the emission (especially the first documents) contained some contradictions.

Ultimately, the collective work of the experts and fruitful discussions led to the development of a base estimate of the composition, activity, and dynamics of emission. These estimates played the main role in the development of programs for containing the consequences of the Chernobyl accident. To a large degree these estimates were based on experimental investigations of the yield of fission products from overheated nuclear fuel, which were previously performed at the I. V. Kurchatov Institute of Atomic Energy [9]. Being a subject of discussion, these data still were never seriously re-examined in later investigations [9–13].

Method of Analysis of the Emission of Radioactive Substances. Three different approaches were used to solve the problem:

estimation of the activity of the emitted substances according to single measurements of the exposure dose rate for γ-radiation near the power plant;

forecast of the dynamics of the emissions, produced as a result of the accident, according to a model of the yield of fission products during the burning-off of the irradiated fuel, specifically, uranium-graphite fuel elements of a high-temperature gas-cooled reactor;

Russian Science Center "Kurchatov Institute." Translated from Atomnaya Énergiya, Vol. 78, No. 6, pp. 403–417, June, 1995. Original article submitted February 21, 1995.

1063-4258/95/7806-0390$12.50

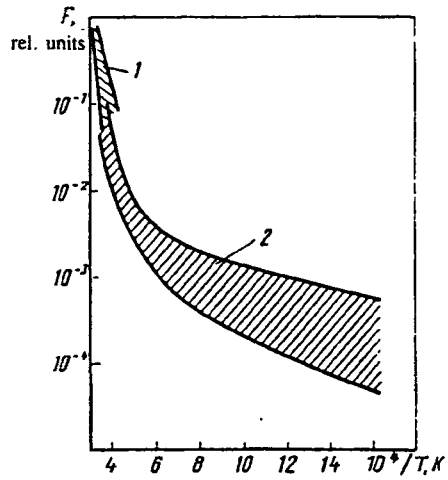

Fig. 1. Relative leakage of ^{85}Kr, ^{137}Cs (1), and ^{131}I (2) as a function of the temperature with short-time burnup (τ ~ 1–2 h).

determination of the fraction of emitted radionuclides and fuel from systematic local measurements of the exposure dose rate and the spectral composition of the radiation.

In the analysis of the primary information on the radiation conditions, the data obtained prior to May 6, 1986 were taken as the most reliable data. This is because the assumption that the washing out, dissolution, and oxidation of UO_2 and other physical-chemical processes as well as housekeeping work could change the ratio of the radionuclides in the soil and the dose fields caused by them. This hypothesis was later confirmed by many experiments on natural materials.

The activity on the l-th day was determined from the relation $Q = \sum_l \sum_m K_{ml} P_{ml} S_{ml}$, where K_{ml} is the coupling coefficient between the exposure dose rate and the specific activity in the m-th region, Ci·km^{-2}/R·h^{-1}; P_{ml} is the average exposure dose rate measured in the m-th region, R/h; and, S_{ml} is the area of the region with an average exposure dose rate of P_{ml}, km^2.

The rate of emission of radionuclides R(Ci/day) was estimated from the daily activity of the emissions, which was found from the relation $R = Q_l - Q_{l-1}$, where Q_l and Q_{l-1} are the total activity of emission over the l-days after the accident and over the preceding l - 1 days.

The coupling coefficient K_{ml} was determined using the expression for the exposure dose rate above the half-space with a uniform density of surface radioactive contamination, containing j radionuclides:

$$P_{ml} = \sum_{i,j} \frac{Q_{mj} n_{ij} E_i}{2k_\gamma (E_i)} \Phi(\mu_i h) = Q_m / K_{ml}.$$

Here Q_{mj} is the activity of the fallout of the j-th radionuclide in the m-th region; n_{ij} is the yield of γ-rays with energy E_i per decay of the j-th radionuclide; the function $\Phi(\mu_i h)$ characterizes the absorbing properties of the material between the radiation detector and the source; and, $k_\gamma(E_i)$ is the coupling coefficient between the flux of γ-rays with energy E_i, MeV/(cm^2·s), and the dose rate produced, R/h.

The results of investigations, performed previously at the I. V. Kurchatov Institute of Atomic Energy, of the leakage of fission products during heating of UO_2 and uranium-graphite fuel elements of a high-temperature gas-cooled reactor [9] were used as the base data on the yield of the fission products. Specifically, these experiments established the following dependence of the degree of volatility of the elements on the temperature T:

Xe, Kr – most volatile radioactive inert gases;

I, Te – volatile in almost the entire range of temperatures T for the characteristic for the accident, i.e. for $T >$ 100–200°C;

Cs – volatile at temperatures $T >$ 800–1000°C; as the temperature increases, its volatility approaches that of iodine and the radioactive inert gases;

Ba, Sr – volatility close to but less than that of cesium (barium is more volatile than strontium);

TABLE 1. Fractionation Coupling Constant K_f and the Characteristics of Emission of Radionuclides and Fuel

K_f	Carrier	Temperature, °C	Emission mechanism	Emission and fallout zone
~1 ± 0.2:				
Holds for all elements	Fuel particles	$T \sim T_{work}$	Mechanical processes	Any, near the plant
Does not hold for all elements	Free radionuclides (gas phase, aerosols)	$T_{ef} > T_{melt}$	SCR, dissociation of fuel	Most of the fuel and fission products far from the plant
Holds for refractory, nonvolatile elements		$T > T_{work}$	Explosion followed by heating of the fuel	Partial emission of fuel and fission products
<1: for I, Cs, Xe, Kr	Fuel particles			
>1:				
for I, Cs	Fuel particles + free radionuclides (gas phase, aerosols)			
if $K_f(I) > K_f(Cs)$		$T < 1800$		
if $K_f(I) \sim K_f(Cs)$		$T > 1800$		

Note: T_{melt} – melting temperature of the fuel; T_{work} – temperature of the fuel at nominal reactor power; T_{eff} – average temperature of the fuel in a reactor corresponding to the measured leakage; SCR – spontaneous chain reaction.

the remaining elements are volatile at temperatures $T > 1700$°C (Ce, Zr, and Nb are least volatile); and,

Pu, U – plutonium is slightly more volatile than uranium but less volatile than cerium.

The phenomenological activation model previously developed at the I. V. Kurchatov Institute of Atomic Energy was used to analyze the behavior of the fission products in the fuel and the structural materials. According to this model, the leakage r_i from the fuel matrix depends only on the decay constant of the i-th radionuclide: $r_i = \text{const}\ (\lambda)^{-n}$, where λ_i is the decay constant and $0 < n < 0.5$.

The activity of zirconium of constructional origin (~5% of the activity of zirconium – a fission product) was also taken into account in determining the activity of ^{95}Zr and ^{95}Nb in the emission. Depending on the ratio of the emitted quantity of fuel and structural zirconium, this could increase the ^{95}Zr activity in the fallout by a factor of 1.5–2, as subsequent analysis of samples confirmed.

Basic Quantitative Parameters. It was determined that for the first phase of the accident with ^{131}I excess, which is characteristic for this phase, the coupling coefficient $K_{ml} = 1$ R/h/10^5 Ci/km^2 and at the end of the period analyzed $K_{ml} = 1$ R/h/$2 \cdot 10^5$ Ci/km^2. It was shown that for the energy range of importance in practice $0.1 < E_i < 2$ MeV, the coupling coefficient $K_\gamma(E_i) = 5.5 \cdot 10^5$ MeV·s^{-1}·cm^{-2}/R·h^{-1}.

The fractionation coefficient $K_f = (A_i/A_{ref})^{exp}/(A_i/A_{ref})^{calc}$, which characterizes the relative enrichment or depletion of a sample with respect to the radionuclide under study as compared to the average content of the radionuclide in the core, was used as one of the basic quantitative parameters. Here A_i is the activity of the i-th radionuclide (fission product or actinide) in the sample, as determined experimentally (the index "exp") or calculated at the end of the production period of the reactor (index "calc") taking into account the radioactive decay at the moment of measurement and the average over the volume of the core, where the fuel elements with different burnup were present; A_{ref} is the activity of the reference (standard) radionuclide. The least volatile radionuclides ^{141}Ce and ^{144}C were used, as a rule, as the references.

The value of K_f yields information about a process which is characteristic for emission of radionuclides and fuel from a reactor (Table 1). Moreover, this quantity can be used to estimate the effective temperature T_{eff} of the emitted fuel. The average temperature of the fuel in the reactor of which the emission of fission products is equal to the measured emission was taken as the effective temperature T_{eff}. If $K_f(I, Cs) < 1$, then the short-time relative volatility F of a volatile radionuclide is $F = 1 - K_f(I, Cs)$, and T_{eff} can be estimated from its temperature dependence, determined in the experiments of [9] on the heating of irradiated fuel (Fig. 1). For $K_f(I, Cs) > 1$ this parameter must be averaged over its characteristic values for all the main fallout zones of the fission products, and the value obtained must be used to estimate T_{eff} for a medium in the reactor unit from which the fission products emerged. Information about the coefficient F was used at the first stage of the accident to estimate T_{eff} as well as the amount of fuel, plutonium and other actinides, emitted from the damaged power-generating unit.

TABLE 2. Relative Fraction of Activity of Plutonium Isotopes*

Degree of burnup, MW·days/tonnes	^{239}Pu	^{240}Pu	^{241}Pu	^{238}Pu	Activity, Ci	^{144}Ce
10300	0,29	0,38	$2\cdot10^{-3}$	0,32	$8{,}5\cdot10^{4}$	$1\cdot10^{-3}$
14385	0,235	0,38	$1{,}9\cdot10^{-3}$	0,39	$1{,}2\cdot10^{5}$	$1{,}4\cdot10^{-3}$
8300	0,41	0,365	$1{,}58\cdot10^{-3}$	0,21	$6{,}2\cdot10^{4}$	$7{,}2\cdot10^{-4}$
4800	0,59	0,31	$8{,}75\cdot10^{-4}$	0,1	$3{,}2\cdot10^{4}$	$3{,}7\cdot10^{-4}$
900	0,88	0,11	$1\cdot10^{-4}$	0,1	$5{,}32\cdot10^{3}$	$6{,}2\cdot10^{-5}$

*Results presented by E. V. Burlakov (July 1986).

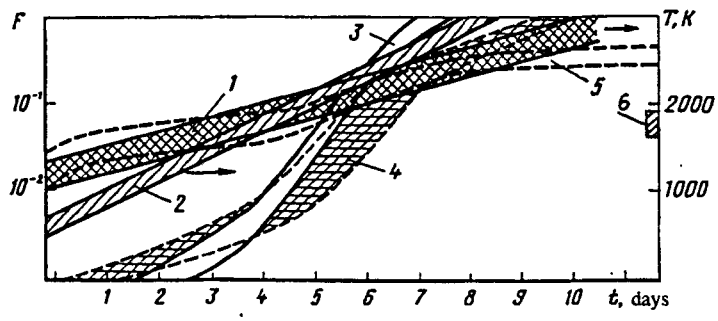

Fig. 2

To estimate the amount of fuel and plutonium emitted, we employed the experimental data on the activity of the least volatile radionuclides (^{141}Ce, ^{144}Ce) in local samples of the fallout as well as the computational data, presented by E. V. Burlakov in July 1986, on the amount of plutonium accumulated in the reactor (Table 2).

Since continuous reloading of fuel is used in the RBMK reactor and the fuel assemblies in the reactor have different burnups, a wide spectrum of ratios of the different radionuclides is observed in the samples. As a rule, during emission from the reactor, averaging of the concentration of the radionuclides occurred and the concentration approached the composition characteristic for the average production (10.3 MW/kg over a period of 640 days). Here the expected chemical forms and the features of the process of separation of the fission products from the overheated nuclear fuel had to be taken into account. Specifically, at ~2300 K volatile fission products predominate in the samples and at temperatures >2300–2400 K the ratio of the radionuclides shifts toward the ratio characteristic for fuel with the average properties. It is important that emission of fuel particles containing besides uranium and plutonium, the minimally volatile elements, specifically, cerium, could also have been emitted. For this reason, to determine the amount of plutonium in the samples, besides laborious measurements of the α-activity of plutonium, data on the γ-activity of cerium was also used. Before July 1986 the ratio of the α-activity of plutonium and cerium in fuel with the average composition was taken to be $9\cdot10^{-4}$. This differs very little from $1\cdot10^{-3}$ for the average fuel burnup, as presented in Table 2.

At the earliest stage of the accident it was also noted that the separation of fission products occurring due to the dissemination of the radionuclides themselves and their carriers over the soil surface and causing the fallout composition to deviate from the average statistical characteristics of the emission must be taken into account in the analysis of the fallout samples. For this reason, separate samples could have properties which are characteristic of different microzones of the fuel and other fuel-containing media, specifically, a high content of volatile and nonvolatile radionuclides (associates), in particular, in the analysis of fuel and hot particles. Moreover, the isotopic ratios between the actinides and other elements could also break down. These features underscored the necessity of performing a mass analysis of samples of different media with high specific activity in order to obtain a correct estimate of the emission during the accident.

Four Stages of Radioactive Emission. During the first few days of the accident the initial data on the radiation conditions near the power plant consisted of only the following three quantities:

TABLE 3. Radiation Conditions near the Chernobyl Nuclear Power Plant and Estimates of the Activity, Emission Intensity, and Leakage of ^{131}I [2, 3]

Date	Exposure dose rate in the zone, R/h	Area of the zone, km^2	Activity in the zone, MCi	Relative leakage of ^{131}I, %*
May				
02	$1{,}2 \cdot 10^{-1}$	38	0,45 } ~ 2	
03	$1{,}5 \cdot 10^{-2}$	490	1,5 }	
	$4{,}5 \cdot 10^{-1}$	1150	5,2	9,2
	$2{,}0 \cdot 10^{-1}$	54	1,0 } 9,2	
	$7{,}5 \cdot 10^{-3}$	410	3 }	
04	$0{,}2 \cdot 10^{-3}$—$4{,}0 \cdot 10^{-3}$	$1{,}6 \cdot 10^{3}$	16—19	
05	$2 \cdot 10^{-2}$—$1{,}6 \cdot 10^{-1}$	$4{,}4 \cdot 10^{3}$	20	13,2
06	$1 \cdot 10^{-2}$—0,5	$5{,}5 \cdot 10^{3}$	29	19,9
07	$7 \cdot 10^{-3}$—0,15	$5{,}0 \cdot 10^{3}$	16,8	
08	$6 \cdot 10^{-3}$—0,2	$5{,}7 \cdot 10^{3}$	12	
09	$5 \cdot 10^{-3}$—0,6	$4{,}3 \cdot 10^{3}$	14,5	
10	$4 \cdot 10^{-3}$—0,15	$5{,}9 \cdot 10^{3}$	8,7	
11	$6 \cdot 10^{-3}$—0,2	$5{,}7 \cdot 10^{3}$	12,8	
12	$4 \cdot 10^{-3}$—0,15	$5{,}7 \cdot 10^{3}$	10,3	
13	$3{,}5 \cdot 10^{-3}$—0,15	$5{,}7 \cdot 10^{3}$	9,4	

*100% corresponds to the content in RBMK fuel on April 25, 1986.

The results of measurements of the exposure dose rate for γ-radiation at three distances from the damaged power-generating unit. These results were obtained on April 29–30, 1986 by a team of civil-defense specialists near the fourth power-generating unit (300 R/h) and at a distance of 200–250 m from it (10 R/h), as well as by a group from the I. V. Kurchatov Institute of Atomic Energy in a town located 4 km from the power plant (1 R/h);

The results of γ-spectrometric investigations of soil samples from the same town, performed at the I. V. Kurchatov Institute of Atomic Energy. It was found that these samples contained not only fission products characteristic for an operating reactor (the activity of the iodine isotopes was 7–10 times higher and the activity of cesium isotopes was 3–5 times higher than follows from the working data for a RBMK reactor [8]), but also particles of irradiated nuclear fuel in the form of highly active grains of UO_2. An anomalous, never previously observed, composition of radionuclides was also recorded in measuring a smear from an armrest in a bus used to transport firemen and plant workers who were over-irradiated from Chernobyl to Moscow (to hospital No. 6).

The results of analyzes of soil samples performed at the Leningrad nuclear power plant which showed a high fraction of volatile cesium and iodine and almost complete absence of nonvolatile serium and zirconium.

A working hypothesis of emission (May 2, 1986, A. A. Khrulev) was proposed on the basis of this information and previous investigations of leaks of fission products from irradiated fuel heated up to a high temperature [9]. From this analysis it followed that the emission during an accident must be divided into four basic stages:

first stage – emission caused by an explosion due to positive reactivity;

second stage – emission associated with the combustion of the graphite lining of the reactor;

third stage – emission due to processes occurring as the temperature of the fuel and fuel-containing mass increases as a result of the energy of radioactive decay of the accumulated fission products; and,

fourth stage – sharp decrease of the emission as a result of stabilization and subsequent gradual decrease of the temperature.

Different measures taken in the damaged power-generating unit could have influenced the dynamics of emission, depending on the effectiveness of the measures.

The following assumptions, which were made on the basis of the first experimental data and hypotheses, were most important:

TABLE 4. Activity of Emitted Substances and Distribution of Fission Products in the Damaged Power-Generating Unit and Outside It at the First Stage and at the End of the Third Stage [5]

Radionuclides	First stage (April 26)		Activity of radionuclides on May 6, 1986 (3rd stage)						
	Activity of emitted substances		Inside unit				Total, %	Outside power-generating unit	
			In fuel		In the fill and the structures				
	Absolute, MCi	Relative, %*	Absolute, MCi	Relative, %	Absolute, MCi	Relative, %		Absolute, MCi	Relative, %*
Radioactive inert gases:	10	<3	9	<20	1	2—3	<20	36	<80
Including									
^{133}Xe	5	<4	9	<20	1	2—3	<20	35	<80
^{85m}Kr	0,15	1,5	—	—	—	—	—	—	<80
^{85}Kr	—	—	0,2	<20	0,015	2—3	<20	0,7	<80
Volatile fission products (except cesium)	15	<2	<20	<20	<63	<63	83	17	17
Including									
^{131}I	4,5	5	7,3	<20	22	<60	80	7,3	20
^{132}Te	4	4	2,4	<20	7,8	<65	85	1,8	15
Cesium:	1	1	4,6	<20	16	<70	90	2,3	10
Including									
^{134}Cs	0,15	1	1	<20	3,5	<70	90	0,5	10
^{136}Cs			2,0	<20	7,2	<72	92	0,8	8
^{137}Cs	0,3	1	1,6	<20	5,4	<67	87	1,0	13
Nonvolatile fission products	8—10	0,3—0,4	300—600	20—40	800—1100	57—78	<97	30	2—3
Including									
^{90}Sr	0,016—0,02	0,3—0,4	1—1,5	20—30	3,7—4,2	66—76	<96	0,2	4
Actinides	0,25—0,3	0,3—0,4	70	<80	15—16	17—18	97—98	1,7—2,5	2—3

*100% corresponds to the activity of radionuclides in RBMK reactor on April 26, 1986.

during the explosion (first stage) the emission of fuel heated up to a high temperature occurred in a solid phase, almost without melting of UO_2;

the high fraction of volatile radionuclides indicates that relatively little fuel was emitted; and

at the third stage, depending on the rate of heating of the damaged reactor (5 or 10°C/h), a repeated increased emission, which could have been expected on May 5–10, 1986, can be observed. The predicted dynamics of iodine emission from the damaged reactor, performed on May 1, is compared in Fig. 2 with the results of field measurements. The radiation conditions near the power plant, which were estimated from the civil-defense maps, and the actual conditions are presented in Table 3.

At the first stage (April 26, 1986) the emission of radionuclides was determined by processes associated with positive reactivity and short-time increase of the effective temperature T_{eff} of the fuel up to $\sim 1600-1800$ K, followed by rapid cooling of the fuel to the temperature of the graphite brickwork. The later refinement of the integral emission of cesium and also the experimental results on the burning off of fuel required that T_{eff} be increased up to 1800–2000 K.

At the beginning of the first stage the fuel assemblies were destroyed and fuel was dispersed as a result of thermal stresses arising during the heating of the fuel and also expansion of the accumulated fission products in the closed pores of the fuel (gas "explosion"); almost immediately after this a second, steam explosion occurred under the action of, specifically, the heated dispersed fuel which entered the coolant.

At the first stage fission products were transported mainly in the form of gas and aerosol, as well as particles of fuel with different dispersity. Near the power plant part of the fuel in the fallout (plant zone, buildings of the power plant) could have been emitted in the form of coarsely dispersed particles and parts of tablets and fuel assemblies in an explosion. In May 1986 it was assumed that fuel (uranium, plutonium) can be present at a large distance from the nuclear power plant in fallout mainly in the form of particles dispersed up to the sizes of the UO_2 grains, and a high fraction of volatile iodine and cesium should be observed. (Such sections were later found and were termed cesium spots.) It was assumed on the basis of the data presented in Table 4 that at this stage a relatively small fraction of the fuel ($< 1\%$) was emitted from the reactor outside the confines of the plant site. (Later field data on the emissions, especially in foreign countries, required that this fraction be increased.)

For the second stage (April 26 to May 1, 1986), determined by the combustion of the graphite and the initiation of burning of the fuel after the initial effective temperature T_{eff} decreased, it was assumed that finely-dispersed particles of fuel, embedded in the graphite, as well as fission products, embedded in the graphite and sorbed by the graphite during the explosion of the core, are emitted together with the products of combustion. (It was determined later that at the stage of graphite combustion the rate of decrease of emission, neglecting the active measures, can be extrapolated by an exponential law with a constant $\sim 0.5\ day^{-1}$. This confirmed the lower, compared to April 26, 1986, temperature of the fuel.)

At the third stage (May 2–6, 1986) the increase in the emission of radionuclides was determined mainly by the heating of the fuel as a result of radioactive decay of fission products up to $T \sim$ 2500-2800 K, exceeding the initial temperature (April 26, 1986). It was conjectured that at first the fraction of volatile radionuclides probably increases rapidly. At the end of this stage, at the highest temperature, some leakage of nonvolatile Zr, Nb, and Ce (together with the fuel) in addition to the volatile radionuclides can occur. It can be assumed that almost all of the radioactive inert gases I, Te, and Cs escape from the fuel and from the power-generating unit, and only part of the remaining radionuclides escaped (see Table 4). After escaping from the fuel, a large fraction of the fission products settled on the materials located in the reactor unit and on the structures of the core and the fill dropped from helicopters into the building containing the damaged power-generating unit. It was conjectured that during this period the fill decreased the iodine emission by a factor of 1.5–2 and the cesium emission by a factor of 2–2.5. (Analysis of the state of the fill in the damaged unit, performed in the last few years, did not definitely confirm this conjecture.)

The last (fourth) stage (after May 6, 1986) must have occurred after stabilization, with the temperature of the main fuel mass of the damaged reactor decreasing gradually and the emission of radionuclides decreasing sharply. This hypothesis was based on the activation model and experimental results obtained at the I. V. Kurchatov Institute of Atomic Energy long before the accident. These experimental results showed that the rate of leakage of fission products during heating of fuel can be 10^2-10^4 times higher than in the case of a stable temperature or decreasing temperature.

TABLE 5. Activity of Radionuclides in the Fallout in Separate Regions (estimated from measurements performed during the first few days of May 1986 [2, 3]), MCi

Region	Date of measurement				Fallout density, Ci/km^2
	10.05	11.05	12.05	13.05	
Belarus–Western Ukraine	18,4	14,5	12,2	16,0	
Including					
Kiev–Mozyr'	14,5	10,5	8,5	11,5	20—600
Gomel'–Mogilev	0,6	1,3	0,7	1,2	20—70
Pinsk–Brest	0,8	0,4	0,5	0,4	
Central Nonchernozem zone–Eastern Ukraine–Povolzh'e	3,5	3,0	2,0	2,5	
Including					
Kaluga–Tula–Astrakhan'	2	1,4	0,5	0,9	20—50
Caucauses					
Including					
Batumi–Tuapse			1	1	Up to 20
mineral waters			0,12	0,1	Up to 6
Armenia				0,01	Up to 3
Baltic Region			0,2		
Leningrad–Tallinn			0,2	0,15	
Murmansk				0,03	

Experts refined the characteristics of the stages and dynamics of emission throughout the entire period of development of the accident. This analysis was based on the results of dosimetric measurements on the territory of the power plant and the adjoining areas and determination of the radionuclide composition of the emission from the γ-spectrometric investigations of air and fallout samples. Aerosol samples were obtained on filters above the reactor with the aid of helicopters and soil samples were obtained at the location in accordance with the wind rose, characteristic for different stages, from zones with the smallest distortions resulting from the superimposition of the emission. It was conjectured that this distortion is smallest in the zone of maximum deposits along the wake. (For distant samples it could be expected that the relative fraction of the volatile component increases as a result of fractionation of the fission products during transport over a long period of time.) Specifically, for the first stage of emission the data for the main wake (town of Pripyat', region of the "rust-colored" forest) and the distant fallout (Great Britain, Sweden, Finland, Leningrad nuclear power plant, town of Plavsk) were found to be preferable. For the second stage the aerosol soil samples collected in the southern and south-western directions were preferable.

It was conjectured that during the first two stages the thermochemical oxidation and carbonization of UO_2, resulting in the separation of fission products from the fuel, do not play a determining role – during the first stage because of the competing oxidation of the graphite brickwork and zirconium under conditions of oxygen deficiency and during the second stage because of the relatively low temperature of the fuel.

At the third stage γ-spectrometric analyses of aerosol samples gave the most reliable information. It was recommended, on the basis of the direction of the winds preceding this period, that the fallout samples be collected in regions which were located outside the wake which precipitated on April 26, 1986.

The civil-defense maps containing the measurements of the exposure dose rate for γ-radiation near the power plant were used to estimate the activity and dynamics of emission at this stage. These data (see Table 3) confirmed the presence of a second emission whose activity level was higher than that of the first emission.

The first maps from γ-aerosurveys, performed by the State Committee on Hydrometeorology, appeared at the I. V. Kurchatov Institute of Atomic Energy after May 6, 1986. These maps made it possible to integrate the fallout over the contaminated territory and estimate the activity of the radionuclides which precipitated in different regions of the country (Table 5). During this period the results of analyses were obtained from Finland, Sweden, and Great Britain and gave additional information about fallout abroad, and the first analyzes of the composition of the fallout outside the confines of the zone of the power plant were performed (in the towns of Bragin, Radin, Plavsk, and others), the first hot particles were separated by the flotation method and their composition was analyzed, and the residual ^{85}Kr content in the dispersed fuel was determined. This made it possible to determine more accurately the fuel temperature during the explosion

TABLE 6. Relative Content of Radionuclides according to Results of γ-Spectrometric Analysis of Aerosol Samples Collected on a Filter above the Fourth Power-Generating Unit in April–May 1986 (relative to ^{144}Ce activity, numerator – data of [11], denominator – [5]; the values for a different sample collected on the same day are indicated in parentheses)

Date	^{95}Zr	^{103}Ru	^{106}Ru	^{132}I	^{132}Te	^{134}Cs	^{137}Cs	^{140}Ba	^{141}Ce	Total activity, Bq/m^3	Remark
April											
28th	1,1	1,2	1,9	4,7		2,1	1,3			$\sim 1{,}5 \cdot 10^8$	Emission of radionuclides associated mainly with combustion of graphite
29th	2,1	1,3/1,02	3/1,8	3,5/4,25	9,6	2,5/5,5	6/3,4	1,6	0,6	$7{,}1 \cdot 10^7$	
May											
1st	0,9	0,85	1,8	22		10	7				Reactor temperature higher than the temperature at the first stage of the accident
2nd	—/1,3	0,8,/0,6	1,8/0,7	26/2	—/3,8	10/2,1	7/30	1,4	0,92	10^7	
3rd	0,9/0,5	0,8/6,1	1,6/12	10/52	/70	10/31	8/44	/3,9	/0,7	$3 \cdot 10^7$	
4th	0,9/1,1	1/1,2	1,6/0,95	4/4,1	/6,8	2,3/2,1	2,1/2,7	/3,2	/1,0	$8 \cdot 10^7$	Increase of the temperature is completed
5th	0,9/1,5	0,9/1,2	5,5/3,3	25/2,9	/6,2	10/	6/2,6	/1,6	/1,1	$1 \cdot 10^6$	High ruthenium oxides appeared – indicating onset of intense oxidation of UO_2
7th	0,9/	3,5/	7,2/	8/	/	10/	6/				
8th	0,9/	12(23)/	18(3,5)/	15(110)/	50/	10(9,1)/	6(28)/	2,8/	1,3/	$1{,}5 \cdot 10^4$	
9th	0,9/1,02	1/4,8	2/	10/30	/50	10/	7/	/3	/0,8	$3 \cdot 10^5$	
10th	0,9/	1/	2/	9/		10/	7/				
11th	—/1,07			/25		—/5,3	—/3,4		—/0,91	$2 \cdot 10^5$	
12th	—/0,95	—/10		—/66					—/0,85		
13th	—/1,04	—/2,3		—/12			—/12			$6 \cdot 10^3$	
14th	1	1(1,3)	1,8(0,7)	5(4,8)	(30)/	70(1,8)	90(2,6)			$3 \cdot 10^3$	
15th	1(1)	0,05(1)	0,1(1,5)	0,3(10)		3,3(1,7)	3,5(2,8)	(0,9)	(1,4)	(80)	
16th	1,1(1)	0,7(0,8)	0,9(0,8)	0,6(2)		4(0,5)	5(0,8)	(0,7)	1(1)	(10^4)	
17th	9(1,9)	1(0,7)	1,5(0,6)	7(6,4)		10(0,75)	8(0,95)	(0,35)	(1)	$(2 \cdot 10^2)$	
18th	1/3,7	1/18,5	10	7/135		50	50				
19th	2(1,1)	1,2(6,2)	1,6(5,4)	8(41)		10(1,6)	7(2,7)	(0,5)	(1)	(500)	
20th	1	1,2	3,6	8		10	7				
24th	—/1,4	—/5,5		—/23		—/2,0	—/3,0		—/0,7		

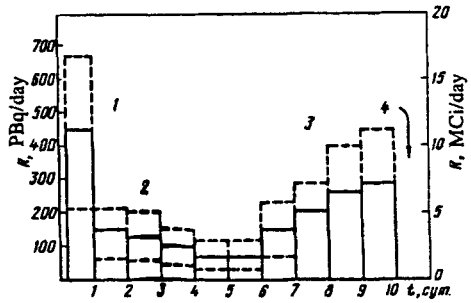

Fig. 3. Intensity of radionuclide emission from the damaged power-generating unit of the nuclear power plant [7]: 1 – initial emission; 2, 3 – period of cooldown and heating, respectively; 4 – sharp drop (down to 2–6 PBq/day); --- – range of uncertainty.

(G. V. Momot and A. A. Khrulev). It correlated with the initial assumptions. Moreover, the possibility of determining the quantity of fuel from measurements of the dose fields and the region of applicability of this method were assessed. By this method it was possible to identify 5–10 tonnes UO_2 in the zone of the reactor (A. V. Khrustalev and A. A. Khrulev).

Conservative estimates of possible dose loads receieved by the population were made from data on regional contamination (O. Ya. Shakh). The dose was found to be overestimated, but it showed that steps to protect the population not only in the zone of the power plant but also in other regions of the country had to be taken.

After May 6, 1986 the emission of radionuclides from the damaged power-generating unit dropped sharply and then continued to decrease slowly. During this period the composition of the emission was monitored only from the results of analysis of aerosol samples collected above the damaged reactor and near the nuclear power plant (Table 6). These data showed that individual samples give only a qualitative idea of the radionuclide composition, but not about the emission as a whole. It is important to note that according to these data, tellurium is slightly less volatile than iodine and cesium. Barium and lanthanum, previously assumed to be almost nonvolatile up to quite high temperatures, are also quite volatile. The same is true for ruthenium, whose yield grows especially rapidly with strong oxidation as a result of the appearance of volatile RuO_4. Analysis of the accumulated information made it possible to make a preliminary and then final conclusion that the sharp decrease of the intensity of emission of radionuclides from the destroyed reactor on May 6, 1986 (Fig. 3) is due to the cessation of burning of graphite, completion of the main physical-chemical reactions in the core, stabilization of the temperature, and onset of cooling of the main part of the destroyed reactor.

In accordance with the working hypothesis, it was assumed that additional emissions resulting from heating of the structures on which fission products have settled or for chemical and technological reasons are possible. It was concluded that these additional emissions should not substantially change the total emission and radiation conditions elsewhere on the location, though they can influence the local conditions near the nuclear power plant.

In June 1986 additional estimates of fallout in the near zone of the nuclear plant were performed (B. G. Pologikh, Yu. V. Sivintsev). These estimates refined the information obtained during the first period. From results of numerical integration of the maps of an γ-aerosurvey of the country and sporadic information about the density of fallout abroad, results concerning the values of the absolute (50 MCi) and relative (3.5% of the accumulated activity) emission of radionuclides from the damaged power-generating unit (neglecting the emission of radioactive inert gases), presented by May 6, 1986 (Yu. V. Sivintsev, A. A. Khrulev, O. Ya. Shakh), as well as data from a quantitative estimate of the fuel distribution in the damaged reactor (with 50% error [4, 5]), were presented to the government commission, working in Chernobyl, and the Politburo of the Central Committee.

In June 1986 the analysis of the composition of the radioactive fallout in different regions of the country continued. The first experiments on estimating the temperature and character of the interaction of the fuel with concrete and soil as well as the temperature dependence of the cesium confinement by the fill (V. P. Avdeev, A. D. Poyukhov, G. V. Momot, I. V. Zakrzhevskaya, A. A. Khrulev) were performed. A simplified physical model was developed for the heating of the fuel taking into account leakage and energy transfer by fission products, and calculations of the dynamics of emission and the fuel temperature were performed (A. A. Vedenov, O. P. Ivanov, A. A. Khrulev). These investigations showed that the maximum fuel temperature did not exceed T_{melt} and increased up to 2750–2900 K only briefly. On this basis it was predicted that destruction of the main building should not be expected ("destruction not obvious").

TABLE 7. Estimate (with an error of 50%) of the Radionuclide Composition of the Emission as a Result of the Accident [6, 7] (100% corresponds to the activity of the radionuclide in RBMK fuel on April 25, 1986)

Radionuclide*	Activity of emission		
	Absolute, MCi		Relative, %
	26.04.86	06.05.86**	
^{133}Xe	5	45	Possibly up to 100%
^{85m}Kr	0,15	—	Same
^{85}Kr	—	0,9	"
^{131}I	4,5	7,3	20
^{132}Te	4	1,3	15
^{134}Cs	0,15	0,5	10
^{137}Cs	0,3	1,0	13
^{99}Mo	0,45	0,3	2,3
^{95}Zr	0,45	3,8	3,2
^{103}Ru	0,6	3,2	2,9
^{106}Ru	0,2	0,16	2,9
^{140}Ba	0,5	4,3	5,6
^{141}Ce	0,4	2,8	2,3
^{144}Ce	0,45	2,4	2,8
^{89}Sr	0,25	2,2	4,0
^{90}Sr	0,015	0,22	4,0
^{238}Pu	$0,1 \cdot 10^{-3}$	$0,8 \cdot 10^{-3}$	3,0
^{239}Pu	$0,1 \cdot 10^{-3}$	$0,8 \cdot 10^{-3}$	3,0
^{240}Pu	$0,2 \cdot 10^{-3}$	$1 \cdot 10^{-3}$	3,0
^{241}Pu	0,02	0,14	3,0
^{242}Pu	$0,3 \cdot 10^{-6}$	$2 \cdot 10^{-6}$	3,0
^{242}Cm	$0,3 \cdot 10^{-2}$	$2,1 \cdot 10^{-2}$	3,0
^{239}Np	2,7	1,2	3,2

*Data were obtained from the results of γ-spectrometric analyzes.
**Total emission up to May 6, 1986.

In June 1986 the government commission sent to the Politburo of the Central Committee, a report in which it was suggested that a decision about re-evacuation of populated points be made on the basis of estimates of the internal irradiation dose due to ^{90}Sr, ^{137}Cs, and ^{239}Pu and not only the exposure dose rate due to γ-radiation (Yu. V. Sivintsev, R. F. Razuvanov, and V. I. Matvienko). This document played a decisive role in changing the plans for re-evacuation of a large number of populated points.

In July 1986 data on the activity, composition, and dynamics of radionuclide emission during the accident, which were included in a report presented by Soviet experts to IAEA [7], were finally formulated. The results of the estimates of the composition and activity of the emission from the damaged reactor, which were included in the final document to IAEA, are presented in Fig. 3 and Table 7.

It should be noted especially that the calculations of the composition of radionuclides in the fuel, which were prepared at the end of April 1986 (V. D. Fomenko, A. A. Khrulev), were used in the analysis of the fractionation of radionuclides with normalization with respect to ^{144}Ce prior to the fall of 1986. Comparing these data with the results of more detailed calculations published in 1990 [10] indicates that they agree well with one another (Table 8).

In conclusion it should be noted that in May–July 1986, about twenty documents were released for the computational-experimental substantiation of activity and dynamics of emission at the I. V. Kurchatov Institute of Atomic Energy. Together with the authors of the present paper, the following individuals also participated in the preparation of these documents: V. P. Avdeev, I. V. Zakrzhevskaya, G. V. Momot, V. A. Sysoev, A. F. Usatyi, A. N. Fedosenkov, V. D. Fomenko, and O. Ya. Shakh, whom the authors of the present paper thank for their assistance during those difficult days.

TABLE 8. Relative and Absolute Activity of the Main Dose-Forming Radionuclides Accumulated in Soil at the Moment of the Accident

Radionuclide	K_f, relative units	Activity, PBq	
		according to the data of 1986 [6]	according to the data of 1990
^{89}Sr	0,56	2,1	2,33
^{90}Sr	0,05	0,19	0,23
^{95}Zr	1,25	4,6	4,8
^{99}Mo	1,36	5,1	5,9
^{103}Ru	1,3	4,7	4,8
^{106}Ru	2,8	0,97	0,86
^{131}I	0,83	3,1	3,2
^{132}Te	1,1	4,1	2,7
^{134}Cs	0,05	0,176	0,153
^{137}Cs	0,077	0,28	0,26
^{140}Ba	1,25	4,6	5,0
^{141}Ce	1,47	5,5	5,6
^{144}Ce	1,00	3,7	3,7

LITERATURE CITED

1. Yu. V. Svintsev, A. A. Khrulev, and O. Ya. Shakh, "Estimate of radioactive emission during the accident in the fourth power-generating unit of the Chernobyl nuclear power plant and forecast of the intensity and total activity of emission up to May 14, 1986," Report, Institute of Atomic Energy, May 1986.
2. A. A. Khrulev, "Radiation emissions of fission products on the territory of the country for the period from May 6 to May 13, 1986," Report, Institute of Atomic Energy, May, 1986.
3. A. A. Khrulev and O. Ya. Shakh, "Preliminary analysis of the emission of radioactivity beyond the confines of the damaged power-generating unit at the Chernobyl nuclear power plant and possible long-term radiation consequences," Report, Institute of Atomic Energy, June 1986.
4. Yu. V. Sivintsev, "Estimate of the total activity of the emission of radionuclides from the fourth power-generating unit," Report, Chernobyl, June 1986.
5. "Magnitude and composition of radioactive emission," Report, Institute of Atomic Energy, June 1986.
6. A. A. Khrulev, "Estimate of the magnitude and composition of radioactive emission at the Chernobyl nuclear power plant (main features of the method of analysis)," Report, Institute of Atomic Energy, July 1986.
7. "Accident at the Chernobyl nuclear power plant and its consequences." Information presented by Soviet experts to IAEA, July, 1986.
8. V. M. Kolobashkin, P. M. Rubtov, and V. D. Sidorenko, Radiation Characteristics of the Irradiated Nuclear Fuel [in Russian], Énergoatomizdat, Moscow (1983).
9. N. N. Ponomarev-Stepnoy and A. A. Khroulev, "Release of fission products from high temperature fuel materials," paper presented at the Third International Conference on Thermionic Electric Power Generation, Julich, Germany, 1972.
10. S. T. Belyaev, A. A. Borovoi, E. V. Burlakov, et al., "Reactor fuel for the fourth power-generating unit of the Chernobyl nuclear power plant," Preprint IAÉ-5268, 1990.
11. Yn. L. Dobrunin and P. B. Khramtsov, "Data verification methodology and new data of Chernobyl source term," Radiation Protection Dosimetry, **50**, Nos. 2–4, 307–310 (1993).
12. International Chernobyl Project, Technical Report IAEA, 1992.
13. F. I. Pavlotskaya and B. F. Myasoedov, "Plutonium in soil," Priroda, No. 5, 57–61 (1991).