Technical University of Denmark



Analysis and prognosis of radiation exposure following the accident at the Siberian chemical combine Tomsk-7

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Risø National Laboratory, Roskilde, Denmark October 1994

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Scientific Production Association »Thyphoon«

Risø National Laboratory, Roskilde, Denmark October 1994

Abstract

On the basis of the work (ground investigations and gamma aerial surveys) carried out jointly by the Rosgidromet organizations and Berezovgeologiya, data on the radiation exposure in Russia were obtained shortly after the accident of April 6, 1993 already. These data were transmitted to interested institutions.

The measurements performed on April 11 and 12, 1993 indicated that within the isolines of 10 μ R/h a contaminated area of up to 25 km in length and up to 6 km in width extended towards the northeastern direction. Thus, the contaminated area outside of the premises of the combine covered about 100 km². The total amount of radioactive substances in this area was 530 – 590 Ci. Isotope composition of the radioactive trace was determined by ruthenium-103 (1%), ruthenium-106 (31%), zirconium-95 (22%), niobium-95 (45%) and plutonium-239 (0.02%).

Contamination heterogeneity is caused by the existence of "hot" particles with an activity of up to 10-11 Ci/particle.

In the contaminated area the gamma exposure rate varied between 14 and 42 μ R/h at 1 m height, yielding the maximum external radiation dose 100 mrem/year for the population of Georgievka. The Pu-239 inhalation dose of the population of Georgievka when passing the radioactive cloud did not exceed 1.5 mrem.

A prognosis was made with regard to water contamination of the rivers Samuska and Tom during the flood in spring. Furthermore, contamination of the air layer adjacent to the ground resulting from the wind transport of radionuclides in the summer months at Georgievka was predicted. The values were far below the limits fixed according to the valid radiation protection regulations. However, that radionuclide concentration of the snow water may exceed the limits specified for drinking water.

According to the data measured by the meteorological stations, the radioactive products were not entrained beyond the borders of the country. Source estimation was successfully obtained using RIMPUFF, the Risø on-line puff diffusion model, in backfitting mode.

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Introduction

According to the report about the state of tudiation in the area affected by the accident at the Siberian chemical combine (Tomsk-7), which was submitted by the Commission of the Russian State Committee for States of Emergency [1], the N6102/2 facility of the radiochemical plant was destroyed on April 6, 1993 at 12.58. In this facility, a uranium solution had been prepared for extraction. During the explosion, part of the activity was released into the environment. It can be concluded from the activity data of the uranium solution published by the chemical combine that 500 Ci beta-active and 20 Ci alpha-active products including 19.3 Ci plutonium-239 had been contained in the facility prior to destruction.

Explosion resulted from the decomposition of the organic phase of the solution when interacting with concentrated nitric acid. The limited steam/gas volume released into the hall exploded. Thus, the extent of the damage was increased even further. Activity release into the atmosphere took place via the ventilation system, the pressure and vacuum lines, the 150 m high stack of building no. 205, the ventilation system and stack of building no. 201 as well as via the destroyed walls.

Immediately after Rosgidromet had been notified of the accident, measurements were started and radiation exposure in the area affected was analyzed in accordance with the regulations regarding "The Measures to be Taken by the Rosgidromet Divisions in Case of Nuclear Accidents".

1 Measures Taken by the Russian Federal Survey for Hydrometeorology and Environmental Monitoring After the Accident

At 18.45 local time Rosgidromet was informed about the accident. To determine the contaminated area, all hydrometeorological stations and aeronautical meteorological offices of the monitoring network established by Rosgidromet were ordered to measure the gamma dose rate and the meteorological parameters every hour. The data obtained were then to be made available to Rosgidromet and other interested institutions. These values allowed a preliminary estimation of the extent of contamination to be accomplished. It turned out that contamination was entirely local.

As far as geography is concerned, the contaminated area is slightly hilly with only small differences in altitude (30 m). In the northern part, the territory is mainly covered by coniferous forest with dense underwood. In the south, mixed forest as well as bushes and shrubs are prevailing. Population density amounts to 80%. Heights of 10 to 12 m are reached by the trees. About 20% of the territory is made up of swamps. Nearly 10% is under agricultural use. From east to west, the area is crossed by the Samuska river flowing in strong meanders. A maximum water flow rate of up to 70 m³/s is attained. The mean annual water flow rate is 10 m^3 /s. While in the northeastern part, the territory is mainly loamy (70%), soil \checkmark in the southwestern part is found to be predominantly sandy.

The values measured by the synoptic aerological stations nearby, the meteorological data determined every 30 minutes in Tomsk (table 1.1) and the balloon data measured at Tomsk airport (table 1.2) led to the following conclusions:

	Wind	Wind	Air	Rel.	Preci-	Degree
Time	Dir.	Speed	Temp.	Hum.	pita-	of
	(deg.)	m/s	•C	<u>%</u>	tion	Cloudiness
11:00	190	8-11	- 4.3	79	-	10/3
11:30	200	7-10	~ 4.0	80	-	cirrostratus
12:00	200	8-11	- 3.8	80	-	strato-cumulus clouds
12:30	210	8-11	- 3.5	81	-	-
13:00	190	9 -12	- 3.2	77	-	-
13:30	210	9 -12	- 3.2	77	-	**
14:00	200	10-13	- 2.9	67	-	HT.
14:30	210	9-13	- 2.6	65	-	7
15:00	200	8-12	- 2.3	68	-	
15: 30	200	9-13	- 2.0	71	15 ¹⁴ - 17 ³⁵	•
16:00	200	10-13	- 2.0	81	-	10/4
						cirrostratus
						strato-cumulus clouds

Table 1.1. Data measured by the Tomsk meteorological station on April 6, 1993

Table 1.2. Data measured by the balloons

	altitude above ground (m)													
Time	Grou	nd Level	1	00	2	00	3	00	4	00	5	00	6	00
	dir.	m/s	dir.	m/s	dir.	m/s	dir.	m/s	dir.	m/s	dir.	m/s	dir.	m/s
11:00	200	8	210	7	210	10	220	10	220	14	220	15	220	14
14:00	200	10	200	9	200	10	210	10	210	12	210	15	220	14
17:00	200	3	200	9	200	10	210	10	210	10	-		-	

- Weather conditions at the site of the accident stable southwestern wind (190 210 deg.), speed 8 13 m/s, temperature -3°C; precipitation in the form of wet snow was recorded after the accident at 15:30 local time;
- stratification neutral.

For determining radiation exposure in the area surrounding the combine, two teams of experts of the Institute for Hydrometeorology and Environmental Monitoring, Division West Siberia (ZapSibgidromet), were established. The field measurements were performed at 16 villages and every 300 to 500 m on the roads from Tornsk to Samus and from Tornsk to Nelyubino. The highest contamination was found to exist on the road from Tornsk to Samus. Beginning at km 20, a strong increase in the gamma dose rate was observed. Maximum gamma dose rate was recorded at km 28.6, while high values were still measured up to km 31.

On the basis of the hydrometeorological data and the field measurements, the ZapSibgidromet and Rosgidromet experts were then able to determine the contaminated area within the 30-km zone surrounding the chemical combine. As assumed by the experts, the trace of the radioactive cloud was directed towards the northeast having a length of up to 20 km [2, 3].

Then, the trace dimensions as well as ground contamination inside the "spot" generated had to be determined more precisely. Appropriate methods had already been developed by the Rosgidromet divisions in close cooperation with the Russian Geological Committee when investigating the consequences of the accident at the Chernobyl nuclear power plant. In this case, the measurements were carried out by ZapSibgidromet experts on 7 routes with a total length of 40 km under difficult conditions (deep snow in the taiga). Azimuthal determination of the gamma dose rate was accomplished every 100 m. In addition, 25 snow samples were taken and analyzed with regard to their concentrations of radionuclides and transuranium elements.

On behalf of the Rosgidromet Institute for Global Climate and Ecology and the Russian Academy of Sciences, an aerial gamma survey was made of the 30-km zone surrounding the Tomsk-7 chemical combine. Staff members of the state-owned geological enterprise "Berezovgeologiya" took part in this work [3]. The plane used for this purpose (AN-2) was equipped with a few-channel spectrometer having a Nal(Tl)-8-crystal detector of 200 x 100 mm in size and a total volume of 25 l. This device also served for measuring the gamma dose rate. Sensitivity amounted to 410 pulses per second for 1 μ R/h. Radionuclide composition was determined by recording gamma radiation in the following eight intervals of the energy spectrum: 0.62 - 0.74 MeV, 0.54 - 0.65 MeV, 0.4 - 0.6 MeV, and 0.6 - 0.84 MeV for artificial radionuclides; the measurements for the natural radionuclides took place in the ranges close to the lines of 1.12, 1.76, 1.46 and 2.62 MeV.

Flight altitude was 70 to 100 m. Following the aerial survey measurements, by means of which the general direction of transport and fallout of the radioactive products was found, detailed mapping of the snow-covered contaminated area in the scale of 1:100,000 was achieved on April 12 and 13, 1993 (distance between the measuring routes 1 km). The measuring routes were located at right angles to the axis of the radioactive trace and about 20 km long. The measurements started and ended outside of the trace. Thus, the outlines of the contaminated area could be identified. Calibration of the spectrometer installed in the plane was accomplished by repeated measurements. At the same time, snow samples were taken on the corresponding routes and the dose rate was measured using a dose meter of the type DRG-01. By comparison, a maximum root-mean-square error of the gamma dose rate measurements of 20% was determined.

The results of the gamma aerial survey and the comparative measurements at ground level were analyzed by the scientists involved in the activities and the experts of the Rosgidromet Institute for Global Climate and Ecology. They were found to be in good agreement.

2 Results of the Isotope Analysis of Snow and Soil Samples

From April 7 to April 12, 1993, isotope composition and concentration of the radioactive products deposited on the ground were determined by the ZapSibgidromet experts by means of 27 snow and 10 soil samples. In Fig. 2.1 the sampling is represented schematically. The samples were then analyzed in the laboratories of ZapSibgidromet and SPA "Typhoon", respectively.

Gammaspectrometric analysis was carried out in accordance with the regulations issued by the interauthority commission of the USSR State Committee for Hydrometeorology and Environmental Monitoring (Goskomgidromet). Spectrometers with germanium-lithium detectors and an energy resolution of 3 - 4 keV for the 1336-keV line were applied. The results obtained by the Scientific Production Association "Typhoon" are presented in tables 2.1 and 2.2. The statistical error of the measured data varies between 5 and 25%.

Gammaspectrometric analysis revealed that ruthenium-103 and -106, zirconium-95 and niobium-95 were contained in the samples. By measurement of two snow samples over a period of 8 hours, trace amounts of cerium-144 and antimony-125 (10 - 50 mCi/km²) were detected. Separate gammaspectrometric analysis of the suspended and dissolved fractions of the snow samples resulted in the finding that more than 80% of the activity was bound to the suspended fraction. Gamma spectrometry of molten snow samples without prior separation into a suspended and a dissolved fraction therefore yielded far too high results that could not be used for quantitative estimation of contamination. Nevertheless, the isotope composition and the share of the individual isotopes in total contamination can still be specified.



Figure 2.1. Figure 2.1a shows a schematic represention of the measuring routes, and figure 2.1b a schematic representation of the sampling on April 12, 1993.

Comparison of the values obtained by SPA "Typhoon" and ZapSibgidromet for the suspended and dissolved fraction of the soil and snow samples did not reveal any systematic deviations. However, a scattering factor of 4-5 was reached. Scattering of the data can be explained by the inhomogeneous radioactivity of the samples, which is due to the existence of "hot" particles.

By means of gamma spectrometry of the soil samples, it was found out that the cesium-137 content exceeded gross contamination by a factor of 2-7. The latter had been caused by the emission of cesium-137 during the previous years of operation of the Siberian chemical combine. Existence of this contamination had been confirmed by the investigation results of the ZapSibgidromet experts in the years before the accident.

In numerous snow samples analyzed by SPA "Typhoon", also plutonium-239 and strontium-90 were identified. For plutonium-239 analysis, a method similar to that developed by the V.1. Vernadski Institute for Geochemistry and Analytical Chemistry was applied [4]. Plutonium-242 served as the tracer. The impurity components were removed by an anion exchange resin. Electrolytic separation of

	Radia-	SuspF		Activ	rity, Ci/	km²	
Sample	tion	SolR					
No.	µR/h	Soil	Ru-103	Ru-106	Zr-95	Mb-95	Cs-137
3	70	F	0.0068	1.13	0.843	1.65	
		R	0.0039	0.069	0.022	0.024	
		F+R	0.072	1.2	0.865	1.67	
		Soil	0.026	0.43	0.173	0.347	0.222
		Total	0.098	1.63	1.04	2.02	0.222
4	120	F	0.106	1.89	1.09	2.06	
		R	0.009	0.179	0.05	0.058	
(F+R	1.115	2.07	1.14	2.14	
ļ		Soil	0.223	8.32	6.08	15.6	0.14
		Total	0.338	10.4	7.2	17.7	0.14
7	100	F	0.062	1.46	0.884	1.73	
(·		R	0.003	0.063	0.009	0.016	
		F+R	0.065	1.52	0.893	1.75	
		Soil	0.061	1.36	0.949	1.98	0.435
		Total	0.125	2.88	1.84	3.73	0.435
8	131	F	0.253	4.73	3.51	6.78	
		R	0.015	0.335	0.09	0.137	
		F+R	0.268	5.06	3.60	6.91	
		Soil	0.086	1.99	1.50	3.44	0.271
		Total	0.354	7.05	5.10	10.3	0.271
9	123	F	0.21	4.40	3.24	10.0	
		R	0.006	0.228	0.085	0.311	
1		F+R	0.216	4.63	3.32	10. 3	
5	77	Soil	0.21	4.99	3.34	9.8	0.44

Table 2.1. Snow and soil samples taken in the area affected by the accident (Tomsk-7)

the sample to be counted took place on a stainless steel target. Alpha radiation of plutonium-239 was measured using a spectrometer of the type 570 A-450 R manufactured by the "ORTEC" company. Strontium-90 was determined in accordance with the methods outlined in [5]. They had already been applied in 1990, when SPA "Typhoon" investigated intercalibrated IAEA samples with regard to their plutonium-239, plutonium-240 and strontium-90 contents. Agreement of the results with the basic values was found to be rather good.

The analytical results obtained with regard to the Pu-239 concentration of both suspensions of the particles contained in the snow water of the snow samples studied and filtered water samples are presented in table 2.3. According to the values indicated for the first five samples, at least 90% of the Pu-239 activity of the snow is bound to the water-insoluble, disperse phase of the radioactive fallout. A mean of 4% of the activity is found in the soluble phase only. For subsequent analysis, suspensions filtered from the snow water samples were applied. It is evident from the data given in table 2.3 that the densities of Pu-239 deposition on the snow exceeded 0.06 mCi \times km⁻². However, they were far below the maximum permissible ground contamination (100 mCi \times km⁻²) specified in April 1986 after the Chernobyl accident.

			Radia-	SuspF		Activity,	Ci/km ²	
Route	Sample	Date of	tion	Sol-R				
	No.	sampling	µR/h	Total F+R	<u>Ru-103</u>	Ru-106	Zr-95	Nb-95
M-1	1	12.04.93	30	F	0.021	0.286	0.18	0.465
				R	1		0.02	0.035
				F+R	0.021	0.286	0.20	0.50
M-1	2	12.04.93	23	F	0.061	1.304	0.993	1.97
				R			0.018	0.028
				F+R	0.061	1.304	1.011	2.0
M-2	1	12.04.93	72	F	0.092	2.98	1.51	2.84
(R	ſ	0.05	0.042	0.071
				F+R	0.092	2.13	1.55	2.91
M-3	1	12.04.93	206	F	0.137	3.01	2.11	3.67
				R	0.034	0.843	0. 559	1.036
			_	F+R	0.171	0.385	2.67	4.71
M-4	2	112.04.93	96	F	0.07	1.62	1.01	1.90
				R	0.007	0.154	0.096	0.195
				F+R	0.077	1.77	1.11	2.095
Georgie	evka,	0.7.04.93		F	0.019	0.403	0.224	0.358
village	boundary			R	ļ	0.032	0.013	0.027
field pa	th			F+R	0.019	0.435	0.237	0.385
Georgie	vka	08.04.93		F	0.023	0.566	0.432	0.919
village	boundary			R		0.10	0.046	0.107
house No. 6				F+R	0.023	0.666	0.478	1.03
front ya	rd							

Table 2.2. Snow samples taken in the area affected by the accident (Tomsk-7)

Table 2.3. Density of surface contamination of the snow by plutonium-239

No. or desig-	Anal. fraction	Surface contamina	Pu-239/Zr-95
nation of	Suspension (F)	tion of the snow	activity ratio
the sample	Filtrate (R)	mCi × km ⁻²	%
3	F	0.3	0.036
	R	0.007	
4	F	0.15	0.014
	R	0.017	
7	F	0.4	0.045
	R	0.0032	_
8	F	1.2	0.037
	R	0.043	
9	F	1.2	0.037
	R	0.023	
M-1-1	F	0.06	0.032
M-1-2	F	0.3	0.03
M-2-1	F	0.63	0.042
M-2-2	F	0.35	0.035
M-4-2	F	0.35	0.035
Georgievka,6	F	0.12	0.028

The values given in table 2.3 (last column) are required for the determination of the conversion factors. Using these factors, the Pu-239 content of the radioactive fallout may be calculated from the zirconium-95 activity that can be measured easily. The mean value of the Pu-239/Zr-95 activity ratios indicated in table 2.3 is 3.4 \times 10⁻⁴. The root-mean-square error of the measurements and the mean value amounts to 8 \times 10⁻⁵ and 2.6 \times 10⁻⁵, respectively. From this it may be concluded that the mean value of the ratios above amounts to (3.4 \pm 0.6) 10⁻⁴ with a probability of 0.95. The conversion factor from the zirconium-95 activity measured in a sample to its Pu-239 content ranges from 1.6 \times 10⁻⁴ to 5.2 \times 10⁻⁴. Thus, the most probable surface density at the sampling point of the snow sample M-3-1, the plutonium-239 content of which was not analyzed radiochemically, was found to be about 1 mCi \times km⁻². With a probability of 0.95, the value of 1.4 mCi \times km⁻² was not exceeded.

The snow sample M-3-1 (or to be more precise, the portion analyzed by SPA "Typhoon") was applied for preliminary estimation of the surface density of uranium deposition in the controlled area. According to the data published by the Siberian chemical combine, \$773 kg uranium had been contained in the 6102/2 facility prior to the accident [1]. This value almost completely referred to uranium-238. The fractions of the other uranium isotopes were extremely small. The uranium-238 content of the sample to be investigated was determined by means of neutron activation analysis. It was found that surface density of the uranium-238 deposition on the snow amounted to 480 g x km⁻² at the sampling point of sample M-3-1. Surface density of the alpha-activity resulting from uranium-238 was 0.16 mCi x km⁻². This value may be derived from the known specific alpha-activity of uranium-238 of 3.34 x 10^{-7} Ci x g^{-1} . As the surface density of Pu-239 deposition on the snow amounted to about 1 mCi x km⁻² at this point, the uranium-238/Pu-239 activity ratio of the atmospheric radioactive fallout was 0.16. This value corresponded to that of these radionuclides (0.15) in the source of radioactive emission into the atmosphere. The most important data obtained by the laboratory of the National Health Service with regard to the surface densities of the radioactive Pu-239 and uranium-238 depositions on the snow are reported about in [1] and summarized in table 2.4.

According to the data given in tables 2.3 and 2.4, the surface densities of plutonium-239 deposition on the snow at Georgievka are 0.12 mCi \times km⁻² and 0.20 mCi x km⁻², respectively. Hence, a mean value of 0.16 mCi x km⁻² is obtained. The deviations of the individual values measured from the mean value obviously result from macroscopic and microscopic inhomogeneities of the plutonium-239 deposition field in the Georgievka area. These inhomogeneities may be caused by most of the alpha- and beta-active products being deposited on the surface of the snow in the form of "hot" particles having beta-activities of 10-11 Ci/particle and more. According to preliminary estimates, the deposition density of these particles on the surface of the snow at the fringe of the Georgievka village (field path) amounted to about 4×10^2 particles/m². Now, the special features of ground contamination by "hot" radioactive particles and their physical and nuclear-physical characteristics shall be investigated. According to table 2, values of (6.5 - 7.5) mCi x km⁻² were attained for the surface density of Pu-239 contamination at certain points in the snow. These values corresponded to those of the sections with the highest contamination. But even in these cases, the values were found to be far below the maximum permissible ground contamination by plutonium-239. At measuring points with the gamma dose rate ranging from 160 to 1800 μ R x h^{-1} , a mean value of the Pu-239/zirconium-95 activity ratio of (1.0 ± 0.34) x 10⁻³ was obtained with a probability of 0.95. The individual values were found to be in the range of $(1.0 \pm 0.8) \times 10^{-3}$. Hence, they differed from the above estimates for less contaminated areas in the radioactive trace. It must be pointed out that so

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1			Snow	contamin	ation	
Sampling	Date	Gamma	by alpl	na-active	Total alpha-	
point	1993	exposure	nuclid	es mCixi	km ^{−2}	activity,
		rate $\mu \mathrm{K} \times \mathrm{h}^{-1}$	Pu-239	U-238	U-234	mCi × km ⁻²
	<u> </u>					
Km 28	06.04.	280	0.60	0.151	0.172	0.93
Km 28 left-						
hand side	0.7.04	400	7.51	0.707	0.619	8.94
Km 28 right-						
hand side	07.04	400	5.28	0.815	0.424	6.52
Georgievka	08.04	23	0.20	0.101	0.101	0.40
Km 28						
(50 m)	09.04	370	4.53	0.437	0.306	5.27
Km 28						
(300 m)	9.04	350	2.43	0.334	0.576	3.34
Km 28						
(600 m)	09.04	157	0.59	0.020	0.020	0.65
18 Pl.						
(righ-hand	10.04	1800	6.45	5.15	5.68	18.33 ¹
side of path)	1	ĺ				

Table 2.4. Snow contamination by alpha-active products in the radioactive trace [7].

¹The share of the uranium-235 portion of the sample measured in the total alphaactivity (1.05 mCi \times km⁻² is also taken into account.

far only few measured values have been made available with regard to ground contamination by Pu-239 in the area affected by the accident at the Siberian chemical combine. To obtain more reliable data about the existing contamination, far more measurements have to be carried out.

Thirteen snow samples and 5 soil samples were analyzed by SPA "Typhoon" with a view to determine their strontium-90 content. The results are presented in table 2.5. In the snow, this radionuclide was mainly found in the dissolved fraction, i.e., the snow water. 13% of the strontium-90 was bound to the suspended particles only. According to our data, contamination density of the snow by strontium-90 was the highest in samples nos. 8 and 9. Here, values of 9.2 and 7.6 mCi/km², respectively, were reached. Analysis of seven snow samples by ZapSibgidromet revealed that the strontium 30 concentration of sample No. 1 (18.5 mCi/km²) was twice as high as the maximum value determined by SPA "Typhoon". The other strontium-90 values were in good agreement with the values determined by "Typhoon". Our values do not suggest any relation between the strontium-90 content and the content of gamma emitters in the samples. This can be explained by the fact that the latter are predominantly encountered in the suspended particles of the snow, while strontium-90 is mainly found in the snow water. At the measuring points, density of ground contamination by strontium-90 varied between 140 and 250 mCi/km². These values were much higher than the contamination of the surface of the snow. Gross background radiation (35 mCi/km²) was exceeded by a factor of 4-7. The higher ground contamination may result from emissions during previous operation of the combine. In certain areas, such emissions had been recorded by ZapSibgidromet experts in 1990 already.

Table 2.5. Density of snow and ground contamination by strontium-90, mCi/km²

Sample		Snow		Soil
No.	Snow water	Suspension	Total	
3	2.3			150
4	3.4	0.26	3.6	140
7	2.8	0.35	3.15	210
8	8.5	0.7	9.2	250
9	6.6	1.0	7.6	160
M-1-1	0.3	0.04	0.34	
M-1-2	0.5	0.03	0.53	
M-2-1	0.5	0.07	0.57	
M-2-2	< 0.1	<0.01	0.11	
M-3-1	0.5			
M-4-2	0.4	<0.01	0.41	
Georgievka				
field path	0.55			
Georgievka				
house No. 6	0.6			

3 Spread of the Radioactive Products in the Atmosphere and Contamination of the Ground Due to Their Deposition

On the basis of the total beta-activity values measured for the products contained in the facility, total activity was calculated to be 500 Ci [1]. The activities of 103 Ru, 106 Ru, 95 Zr and 95 Nb are not indicated. With the calibration substance used when measuring the beta-activity of the samples assumed to be 90 Sr + 90 Y (this substance was usually applied at the combine), the activity of the isotopes above can be derived from ratio between their activity and the total beta-activity of the snow samples. The latter was measured using a gamma spectrometer and the RUB-01P beta-system calibrated to 90 Sr + 90 Y.

On April 12, 1993, snow samples were taken by ZapSibgidromet experts along the measuring routes M1 through M4 as well as at the village of Georgievka. As already mentioned above, determination of the total beta-activity and gamma spectrometry of the samples were performed both at the regional ZapSibgidromet laboratory, Novosibirsk and at SPA "Typhoon" in Obninsk. The ratios between the activities of the individual isotopes and the total beta-activity of the snow samples R/β as well as the calculated activities Q_0 of the individual isotopes in the facility exploded during the accident are given below:

Isotope	¹⁰³ Ru	¹⁰³ Ru	⁹⁵ Zr	⁹⁵ Nb	Σ
R/β	0.069	2.3	1.3	3.0	
Q ₀ Ci	35	1150	650	1500	3300

Hence, 3000 - 3500 Ci were contained in the facility prior to the explosion. Assuming that at least one third of the products contained in the facility was released, a release rate of $Q \sim 10^3$ Ci is obtained. As mentioned in the introduction, the roof of building 201 had been partly destroyed by the explosion. On the weather side, explosion had torn a hole of 250 m² in size into the wall. The explosion products were released via this hole, the broken windows, the destroyed roof and the stack of building 201 as well as via the stack of the ventilation system of 150 m in height (building 205) [1]. The individual activities released by each of these sources are not known.

In the Tomsk area and in the 100-km zone surrounding the chemical combine, radioactive contamination is controlled regularly by the measuring and meteorological stations belonging to the ZapSibgidromet radiometrical network. Twenty-three dose control stations, 13 stations controlling radioactive fallout and one station for the control of the concentration of radioactive products in the air (Kolpashevo) are located in the Tomsk region. Following the accident at the chemical combine (Tomsk-7) with the release of radioactive products into the atmosphere, all stations of the ZapSibgidromet radiometrical network in the Tomsk region and in the adjacent areas of Novosibirsk and Kemerovo as well as the measuring stations of the Krasnoyarsk region were ordered in accordance with the valid regulations to carry out measurements of the gamma background until April 11, 1993. From April 6 to April 8, 1993, these measurements were carried out every hour. As of April 9, 1993, they took place every three hours. The gamma exposure rate measured by the measuring stations in the Tomsk region including the control stations in the 100-km zone surrounding the chemical combine was found to vary between 6 and 15 μ R/h on these days. The gamma exposure rates measured in the Tomsk area before and after the accident are presented in table 3.1. It is evident from the data below that gamma background in the 100-km zone did not change after the accident of April 6, 1993. The measuring stations located in the Novosibirsk and Kemerovo areas also did not record any changes of gamma background after April 6, 1993. The mean gamma exposure rates determined by the measuring stations of the radiometrical network from April 6 to April 11, 1993 varied between 7 and 13 μ R/h and 9 and 17 μ R/h in the Novosibirsk and the Kemerovo area, respectively.

The mean values determined by the measuring stations of the Krasnoyarskgidromet from April 6 to April 11, 1993 are presented in table 3.2. For comparison, the mean gamma exposure rates of March 1993 are indicated as well. It can be noticed that the gamma background values measured by the radiometrical measuring stations in the regions adjacent to the Tomsk area did not change after the accident.

The trajectories of air transport calculated for a period of 67 hours following the explosion are represented in Fig. 6.1 for three different altitudes. They indicate that transport took place towards the northern and northeastern direction on the first three days after the accident. However, an increase in the gamma background had not been recorded by any of the Rosgidromet radiometrical network stations located in the direction of these trajectories.

The most sensitive method for controlling the radioactive products released into the air during an accident is the sampling of the atmospheric fallout and aerosols. Radioactivity is measured daily by the stations belonging to the radiometrical network. According to the data transmitted to "Typhoon" by the stations determining the total beta-activity of the samples, a slight increase in the radioactivity concentration of the air and the precipitations was recorded by the Turukhansk station from April 8 to 10 only. On these days, atmospheric precipitation amounted to three times the mean value of the month of March. Concentration of radioactive aerosols was increased by a factor of 1.5 - 2. This, however, was still within the

	Distance of	Azi-	Mean			Apr	il 199	3		
Control	radiation	muth	value							
station	source, km	deg.	1992	1-5	6	7	8	9	10	
Molchanovo	140	325	12	12-13	13	14	15	11	14	11
Kozhevnikovo	65	250	15	15	15	15	15	14	15	15
			1							
Permomaiskoe	100	45	9	9-10	9	10	9	9	10	11
Krasnyi Yar	70	335	7	6-7	8	9	6	6	7	7
		_				••				
Baturino	130	5	11	7-12		12	10	12	_13	14
Beletnee	105	205	10	10	11	11	10	19	11	10
Tomek	105	205		10		11	10	12		10
	30	120	14	12.14	13	12	19	11	19	10
station	50	120	14	15-14	10	10	14	11	12	10
Tomsk	·	- <u> </u>	- <u></u> .							
hvdromet.	20	220	9	8-9	9	10	11	12	12	10
station					•					
Belvi Var	205		19	19_13		0	0	10	11	11

Table 3.1. Gamma exposure rates measured in the Tomsk region before and during the first days after the accident at the chemical combine, $\mu R/h$.

limits of fluctuation of the radioactive background. These increases were of no significance, as precipitations and concentrations of the same amounts had also been observed on some days in March. No radioactivity changes were recorded at the Norilsk station located north of Turukhansk. This means that radioactive contamination had a purely local character. The data measured until April 30, 1993 allow the conclusion to be drawn that no radioactive products were entrained beyond the national borders.

Environmental monitoring after the accident was not only accomplished by the measuring stations, but also by the ZapSibgidromet experts who investigated the area directly near the site of the accident [2]. On April 7, measurements were carried out in the villages of Georgievka and Naumovka with a view to determine the end of the radioactive trace and the extent of contamination. On the next days, measurements were performed along nine footpaths in transverse direction to the trace and along it as well as along the fence surrounding the premises of the chemical combine. Between km 20 and km 31 of the road from Tomsk to Samus, the gamma background was measured on the untouched snow at a distance of 10 to 50 m from the road. The following gamma exposure rates were recorded:

Road section Gamma exposure rate, $\mu R/h$

Km 20-28	12-30
Km 28-28.6	30-510
Km 28.6-28.9	510-180
Km 28.9-29.6	180-200
Km 29.6-31.1	200-20

Measuring station	April 6-1, 1993	March 1993
Bolshaya Murta	11	10
Sukhobuzimskoe	12	11
Dzerzhinskoe	19	17
Kemchug	10	11
Shumikha	11	10
Krasnoyarsk	11	11
Uyar	12	12
Shalinskoe	10	10
Balakhta	16	10
Kacha	10	10
Nazarovo	19	18
Stelka	12	13
Podkamennaya Tunguska	17	13
Yeniseisk	9	17
Achinsk	21	11
Turukhansk	19	19
Motygino	13	13
Vanavara	15	12
Yartsevo	15	17

Table 3.2. Gamma exposure rate measured by the measuring stations of the radiometrical network of the Krasnoyarskyidromet after the accident at the chemical combine, $\mu R/h$.

From these data, the high radioactive contamination between km 28 and km 31 is clearly visible. Therefore, workers of the chemical combine carried out decontamination at km 29 of the road. Thus, the gamma exposure rates in the most contaminated sections could be reduced to $120 \,\mu$ R/h. Along the fence around the combine premises, the gamma exposure rate ranged between 225 and 12 μ R/h from km 28.5 to 30.5. At the guard house at the entry of the combine, a value of 12 μ R/h was measured. At km 28.5 of the road, gamma background measurement was performed up to a distance of 700 m from the road in the direction towards the combine. Gamma exposure rate varied between 180 and 480 μ R/h. Starting at this road section, measurements were made every 500 m until the village of Georgievka was reached. The results obtained are presented below:

Distance, m Gamm	a exposure	rate,	$\mu R/h$
------------------	------------	-------	-----------

up to 270
270-180
180-216
216-240
240-200
200-160
160 -350
350-310
310-280

Of all villages located in the 30-km zone, only Georgievka suffered from contamination. The values measured there are indicated below. At the remaining 14 places (Malinovka, Aleksandrovskoe, kolkhoz "Rassvet", Kopylovo, Kuzovlevo, Bobrovka, Mikhailovka, Nadezhda, Dzerzhinski, Timiryazevo, Zerkaltsevo, Berezovka, Porosino and Nelyubino), gamma dose rate amounted to 7 - 14 μ R/h between April 6 and 12, 1993. At the villages of Karakozovo, Tyukalovo, Yegorovo and Karyukina, a value smaller than 10 μ R/h was determined. At Tomsk-7 and Tomsk, the gamma dose rate was 12 μ R/h which corresponded to the natural gamma background. On the route from Naumovka to Georgievka, the gamma radiation field was found to have a spot-like structure:

3.0 km away from Naumovka,	3 m away from road	150-160 µR/h
	50–100 m away from road	30 µR/h
3.5 km away from Naumovka		17-30 μR/h
1.0 km away from Georgievka,	on the road surface	70 μR/h

After the accident, gamma exposure rate at Georgievka increased to $28-42 \ \mu$ R/h. At certain points on the northern fringe of the village, even values of up to $60 \ \mu$ R/h were recorded. The values measured at Georgievka are represented in Fig. 3.1. Here, the gamma exposure rates are given in μ R/h for certain points of the village. A mean dose rate of $27 \ \mu$ R/h was attained. The gamma exposure rates on the streets were smaller, while on the untouched snow in the surroundings they were found to be very much higher. Here, values of 40 $\ \mu$ R/h were attained. In the fields outside of Georgievka, a value of 30 $\ \mu$ R/h was measured. The measurements on the surface of the ground thus revealed that radioactive contamination of the ground was extremely heterogeneous. This was attributed above all to the existence of hot particles in the aerosol products deposited on the snow.

The results of radioisotopic analysis of two snow samples taken at Georgievka six days after the accident (April 12, 1993) are obvious from table 3.3. According to these data, contamination at Georgievka was mainly caused by ⁹⁵Nb and ¹⁰⁶Ru, while ⁹⁵Zr was of minor importance. Contamination by ¹⁰³Ru could be neglected. All these isotopes are relatively short-lived and, hence, did not appear when determining the composition of the gross radioactive background. They merely represented accident products. Background contamination by ⁹⁰Sr of global origin amounted to about 0.03 Ci/km².

Therefore, concentration of this isotope in the snow could also be neglected. It must be pointed out that ⁹⁰Sr was almost exclusively contained in the aqueous fraction, while the gamma-emitting isotopes were bound to the suspended fractions. 'iotal density of radioactive contamination at Georgievka was 1.6 Ci/km².

The contamination densities of the individual isotopes in the Georgievka area were calculated at a mean gamma dose rate of the entire village of $N_{\gamma\sigma}$ 0=27 μ R/h. They are given in the bottom line of table 3.3. The formulas applied for the calculation are presented in Annex 1. The calculations are based on the assumption of a natural gamma background of $N_{\gamma\sigma} = 10\mu$ R/h. The calculated results are in good agreement with the measured values. Contamination of Georgievka by plutonium (0.1 mCi/km²) can hence be neglected.

On the basis of the data given in table 3.3, external gamma irradiation of the local population may be approximated. For this purpose, it is assumed that no migration of the population takes place. Shielding of the gamma radiation by the walls of the houses and production facilities is neglected. The film contamination of the ground (upper dose value) was calculated using the dose coefficients given in Annex 1 and taking into account the natural isotope migration into the ground.



Figure 3.1. Gamma dose rates at Georgievka on April 12, 1993, µR/h.

Table 3.3. Density of snow contamination by individual radionuclides at Georgievka on April 12, 1993, Ci/km².

Sampling point	Analyzed fraction of the sample	¹⁰³ Ru	¹⁰⁶ Ru	⁹⁵ Zr	⁹⁵ Nb	⁹⁰ Sr	²³⁹ Pu	Σγ
House No. 6	auspension	0.023	0.566	0.432	0.919	0.0000	0.12×10^{-3}	
front yard	water	0.000	0.100	0.046	0.107	0.0006		
	total	0.023	0.666	0.478	1.030	0.0006	0.12×10^{-3}	
Fringe of	suspension	0.019	0.403	0.224	0.358			
the village	water	0.000	0.032	0.013	0.027			
field path	total	0.019	0.435	0.237	0.385			
Mean values calculated							<u> </u>	
according to	total	0.021	0.55	0.36	0.71	0.0006	0.12×10^{-3}	1.6
Eqs. (7) & (12) Annex	total	0.024	0.54	0.37	0.80		0.13×10 ⁻³	1.7

The calculations are obvious from table 3.4 [6]. Reduction of the radiation dose due to the penetration of the isotopes into the soil when digging the gardens and ploughing the fields was not taken into consideration.

Table 3.4. External gamma irradiation of the Georgievka population by the radisactive products released during the accident and the natural gamma background over the period indicated, 10^{-3} rem.

Time, years	Ru-103	Ru-106	Zr-95+ Nb-95	Total	Nat. gamma background
1	0.15-0.16	6.6-8.3	12-14	19-32	70
2	0.15-0.16	8.6-13	12-14	21-27	140
3	0.15-0.16	10-17	12-14		3500

It is evident from the data above that external gamma irradiation of the population is less than 1% of the irradiation resulting from the natural gamma background even when staying permanently (for a period of 50 years) at Georgievka. The calculations were based on the assumptions of $N_{\gamma o} = 10 \ \mu$ R/h and 1R = 0.8 rem (cSv)=0.87 rad (cGy) for air. External irradiation with ¹⁰³Ru, ⁹⁵Zr and ⁹⁵Nb becomes obvious within a period of one year after the accident. Irradiation with the longer-lived ¹⁰⁶Ru is steadily increasing. The shares of gamma irradiation of ¹⁰⁶Ru and ⁹⁵Zr + ⁹⁵Nb in the external irradiation are nearly the same, the contribution of ¹⁰³Ru can be neglected. It may therefore be concluded that external gamma irradiation does not represent any danger to the Georgievka population.

4 Prognosis of Contamination Resulting from Secondary Wind Transport

Judging from the geographical data of the contaminated area, about 10% of the territory is under agricultural use. This territory may be a source of air contamination, when the radionuclides deposited on the fields are transported by the wind. The greatest risk to the population consists in the intake of Pu-239 by inhalation. It therefore seems to be reasonable to estimate air contamination in the area under agricultural use. Air contamination may result from wind or mechanical transport of the deposited radionuclides. Mechanical transport takes place when the soil is cultivated using agricultural equipment or when traffic is passing. Now, air contamination resulting from wind erosion and mechanical impacts shall be estimated. According to the data obtained in the Chernobyl area [7], the intensity of wind transport of recently deposited radionuclides amounts to $10^{-9} m^{-1}$. Assuming that the area under agricultural use is $\sim 10 \text{ km}^2$ with the height of the layer near to the ground surface being 50 zm and the contamination density by gamma and beta emitters ~ 5 Ci/km² (Georgievka), a maximum concentration of these emitters in the air near the ground surface of 5×10^{-16} Ci/l is obtained. This value is smaller than the corresponding dose coefficients DK_B of these radionuclides by four to five orders of magnitude. In our case, air contamination by Pu-239 was 6 \times 10-20 Ci/l (DK_B = 3 \times 10⁻¹⁷ Ci/l). This value was calculated at a contamination density of 8×10^{-4} Ci/km². It allowed the conclusion to be drawn that contamination of the air due to wind transport of the radionuclides was insignificant and did not represent any danger to the population. Mechanical impacts may considerably intensify the wind transport of the radionuclides. The maximum values measured for the intensity of wind transport of Pu-239 are 10^{-4} s^{-1} and 10^{-6} s^{-1} for passing traffic and ploughing of the fields, respectively [8].

Let us now assume that maximum air contamination is caused by public and agricultural traffic passing in transverse direction to the wind. Thus, a stationary active source is generated, the intensity of which may be estimated as follows:

 $Q = I \tau \alpha \rho_s$

where

 $I = \text{traffic density, } s^{-1};$ $\tau = \text{minimum time interval between the individual clouds}$ generated by the traffic; it is calculated using the continuity condition of a jet; $\alpha = \text{intensity of wind transport of the radionuclides, } s^{-1};$

 ρ_s = ground contamination density, Ci/km².

The time τ was calculated using the following equation:

 $\ln(bu_*\tau/ez_0) = \kappa \alpha_{zz}$

where

b	~	0.3 is the universal constant in the similarity theory
		of the Lagrangean turbulence characteristics;
u.	=	dynamic velocity, m/s;
<i>z</i> 0	=	roughness parameter, m;
ĸ	=	von Kármán constant;
e	=	base of natural logarithm;
a ₁₁	=	proportionality factor in the expression for the axial scattering parameter of the impurity.

In the calculations, a value of $\tau \sim 9s$ was obtained at $u_s = 0.2$ and $z_0 = 0.1$ m. With the traffic density being 10 vehicles per hour and the width of the road 10 m, efficiency of the linear source amounts to

 $Q = 2 \times 10^{-13} \mathrm{Ci/m} \times s.$

At a distance of 100 m from the road, Pu-239 concentration then is 2×10^{-18} Ci/l. For comparison with DK_B, this value must be converted into the mean annual concentration. For this purpose, it is now assumed that the dry season, during which the risk of wind transport exists, lasts three months and that traffic is continuous over a period of eight hours a day. Then, a mean annual concentration of 1.5×10^{-19} Ci/l (DK_B = 3×10^{-17} Ci/l) is obtained.

As far as the mean effective equivalent dose was concerned, a value of 0.3 rem was calculated for a period of 50 years. Hence, it did not represent any danger to the population. Estimation of the influence of mechanical cultivation of the soil on the wind transport yields concentration values that are smaller than the above values by two orders of magnitude.

On the basis of the investigations performed, it must be noted that wind transport of the radionuclides does not lead to a dangerous radiation exposure of the population.

5 Prognosis of Water Contamination

5.1 Estimated Radionuclide Concentration of the Samuska River

Following the accident at the chemical combine, also part of the catchment of Samuska river, a subsidiary stream of the Tom river, was exposed to the radioactive fallout. The area covered by the trace was about 100 km². According to the analyses carried out by the experts of SPA "Typhoon", the gamma dose was mainly caused by Ru-103, Ru-106, Zr-95 and Nb-95. As far as the alpha emitters were concerned, Pu-239 was of major importance. Total activity outside of the controlled area of the combine was estimated to range from 300 to 500 Ci. A significant part of the fallout was found to occur in the controlled area. According to the estimations, the total activity released amounted to about 800 to 1000 Ci. For the calculations, a value of 1000 Ci can be used. Then, the activities of the individual radionuclides are as follows:

Ru-103	-	10 Ci		
Ru-106	-	300 Ci		
Zr-95	-	210 Ci		
Nb-95	-	470 Ci		
Pu-239	-	0.1-0.2 Ci		

(Pu-239 fallout was estimated on the basis of the equation:

 $[Pu-239]/[Zr-95] \approx 10^{-3} - 3.5 \times 10^{-4}).$

5.2 Contamination of the flood in spring

The fallout mainly deposited on the snow. Therefore, considerable washout of the radionuclides was expected to take place in the Samuska and Tom rivers when the snow would be melting. The parameters of the flood in spring, which were predicted by the ZapSibgidromet experts, are obvious from table 5.1.

River	catchment area km ²	Surface runoff mm	Flow volume during flood km ³	Mean flow rate m ³ /s	Mean turbidity g/m ³	Specific flow of sediment t/km ²
Tom, Tomsk	67000	946	14	0790	70	
station	37000	240	19	2700	10	
Samuska mouth	505	108	0.054	49	28	0.56
mouth		100	0.001	12	20	0.00
Station near	222					
the combine	333					

Table 5.1. Parameters predicted for the flooding of the Samuska and Tom rivers in spring 1993.

5.3 Estimation of radionuclide washout

Prognosis of the radionuclide flow was based on the homogeneous distribution of the radionuclides in the snow water. While flowing, the radionuclides interact with the earth layer and sorption takes place. According to [9], this interaction layer is about 1 cm thick. It is assumed that a sorption equilibrium exists between the earth and the water flow. This equilibrium is characterized by the distribution coefficient K_d . Only little is known about the distribution coefficients of Ru-103, Ru-106, Zr-95 and Nb-95. According to the data published in [10], however, the behavior of Ru-106 in the soil practically corresponds to that of Cs-137. It was therefore expected that the migration processes of these radionuclides were similar as well. It was shown in [11] that the distribution coefficient of the radionuclides depends on the water/soil volume ratio. Under the flow conditions outlined in table 5.2, this ratio is about 10. This corresponds to a distribution coefficient of $K_d \sim 200$. According to [7], the distribution coefficient of Pu-239 is in the range of $10^3 - 10^4$. For maximization of the estimations, the former value was selected. The washout factors were calculated in accordance with the method described in [12, 13]. It was found out that the washout factors of the dissolved phase amounted to about 5% and 1% for gamma and beta emitting radionuclides and for Pu-239, respectively. In the solid phase, the washout factor does not exceed 0.3% for all radionuclides. It must be pointed out that the estimated washout factors were too high by an order of magnitude at least. This was due to the assumption that all radionuclides were present in the exchange form. It is known, however, that the exchange form is 1 - 5% of the irreversibly sorbed form only [11].

Table 5.2. Mean radionuclide concentration of the rivers Tom and Samuska.

	Radionuclide concentration of the water, pCi/l					
River	Ru-103	Ru-106	Zr-95	Nb-95	Pu-239	
Tom	0.04	1.0	0.7	1.6	0.001	
Samuska	10	150	120	300	0.2	
DK _B , water	80000	12000	62000	96000	2900	

The calculated mean concentrations of the flood are presented in table 5.2. Comparison of the calculated mean values and the permissible concentrations (bottom line in table 4.2) shows that the concentrations in the water are smaller than DKB by a factor of $10^2 - 10^4$ even under the most unfavorable washout conditions. It must be taken into consideration that the limit value DKB was calculated for the annual standardized water consumption with the internal irradiation being 5 x 10-3 Sv.

6 Supply of Information for the Estimation of Radiation Exposure in the Area of the Siberian Chemical Combine

Supply of information for the analysis of radiation exposure in the area affected by the accident depended on the data available and could be divided into two stages:

- A. Analysis of the situation, estimation of the release parameters and determination of preliminary data on the possibly contaminated area and the radiation exposure, estimation of a possible transport beyond the national borders. In this stage, data measured at ground level were not yet available. Therefore, numerous calculations were carried out on the basis of a physico-mathematical simulation of radioactivity spread in the environment using preliminary findings about the source.
- B. Systematization of the data with the aim of setting up a diagram of radioactive contamination of the environment soon after the accident as a function of time and space.

It is the objective of this stage to estimate the statistical reliability of all values measured when investigating the contaminated area and to make use of these data when determining (more precisely) the radionuclide composition of the emissions and their quantitative ratio. Furthermore, a map of contamination in this area shall be prepared (gamma dose rate at ground level, contamination densities of all radionuclides identified in the samples).

As ground measurements were carried out outside of the premises of the chemical combine only, calculations were performed for the entire contaminated area including the combine site. Radioactivity spread in the area as a function of space and time then allowed to estimate the possible individual doses when passing the radioactive cloud.

6.1 Information for the taking of appropriate measures during the first hours after the accident

On April 6, 1993 at about 17.00 local time, SPA "Typhoon" was informed by Rosgidromet about the accident at the chemical combine. For information, Rosgidromet also transmitted the following data on the release:

- Nuclide composition: Pu-239, U-238;
- total activity released: 2 5 Ci;
- activity released into the environment via the destroyed walls of the buildings (height of release up to 30 m);
- duration of the release about 15 min.

On the basis of weather forecasts, possible trajectories were determined for the movement of the radioactive cloud at various heights (ground level, 700 - 800 m and about 1500 m). They are represented in Fig. 6.1.



Figure 6.1.

By simulating the atmospheric transport of Pu-239, radioactivity concentration of the cloud was found to have been decreased to insignificant values due to diffusion and deposition processes within a period of 3.5 hours after the accident. According to the simulation data, maximum Pu-239 concentration of the cloud 3.5 hours after the accident amounted to 1.5E-17 Ci/l (permissible concentration 3.0E-17 Ci/l) at a distance of 110 km from the source and a height of 1 m. Thus, transport of significant radionuclide concentrations (exceeding DKB) beyond the national borders could be excluded.

The values of ground contamination by Pu-239, which were obtained by simulating the atmospheric transport and deposition of the radioactivity, are represented in Fig. 6.3. The gamma aerial survey data obtained later confirmed that the predictions based on the simulation had been very precise.

For simulation, the "Gaussian club" model developed by the data processing center of SPA "Typhoon" according to the method described in [14] was applied. Simulation was performed for a source with a height of 30 m (building, where the accident occurred), a release duration of 15 min. and a total Pu-239 activity released of 5 Ci.

After the receipt of the message, an "express " analysis of the accident was carried out by SPA "Typhoon". The results were then processed and transmitted to Rosgidromet at 20.00 Moscow time on April 6, 1993.

The first data on the radiation exposure were received by SPA "Typhoon" on April 7 and 8, 1993. Gamma dose rates at ground level near the building affected (30 μ R/h), at km 28 of the road from Tomsk to Samus and at Naumovka (14 μ R/h) and Georgievka (40 - 60 μ R/h) were measured by expert teams of ZapSibgidromet and Rosgidromet. On the basis of the first analyses of snow samples, radionuclide composition of the release could be determined more accurately:

Nb-95 - 36%; Ru-106 - 38%; Ru-103 - 1%; Zr-95 - 23%.

The following main source parameters were determined more precisely and recommended for simulation by the Rosgidromet experts:

- Release height 15 150 m (the source was simulated by two simultaneous releases at heights of 15 30 m (50% of the total activity) and 150 200 m (50% of the total activity), respectively);
- deposition rate 0.01 0.19 m/s;
- duration of the release 10 15 min.;
- total activity released 150 400 Ci.

On the basis of the data recommended by the Rosgidromet experts, contamination density of the most important radionuclides was calculated. The map plotted for the contamination density of Nb-95 is shown in Fig. 6.4 (the source was simulated by two simultaneous releases from the building affected (release height 30 m, 50% of the total activity) and the ventilation pipe (height 200 m, 50% of the total activity), respectively, at a deposition rate of the radioactive products of 0.15 m/s and an assumed total Nb-95 activity released of 400 Ci).



Figure 6.2.



Figure 6.9.



Figure 6.4.





 $Ris \sigma - R - 750(EN)$



Figure 6.6. TOMSK - Synthesized map of the distribution of the gamma exposure rate $(\mu R/h)$.

The gamma dose rates at the measuring points (building affected, km 28 of the road from Tomsk to Samus, Georgievka) obtained by simulation at the calculated contamination density amounted to 10 - 20% of the value measured. According to preliminary estimates based on the information available at that moment (April 10, 1993), agreement between the calculations and the measured values could have been achieved with 700 Ci at least. The corresponding calculations were carried out. The results obtained for the Nb-95 contamination density are presented in Fig. 6.4 (the source was simulated by two simultaneous releases from the building affected (release height 30 m, 30% of the total activity) and the ventilation stack (height 200 m, 70% of the total activity), respectively, at a deposition rate of the radioactive products of 0.2 m/s). They were also transmitted to Rosgidromet.

6.2 Systematization of the measured values and data processing for an objective radiation analysis

The following data were processed for an objective analysis of the radiation exposure in the area of the chemical combine [1]:

- Gamma dose rate measured at km 28 of the road from Tomsk to Samus (10 measurements);
- gamma dose rates measured at ground level at different points and along 9 routes in the radioactive trace (about 300 measurements);
- gamma dose rates measured in the villages near the radioactive trace (16 villages);
- radionuclide compositions of the samples determined by the Rosgidromet analyses (11 samples);
- map of the gamma dose rate of the ground; it was prepared on the basis of the gamma aerial surveys performed by the experts of the Rosgidromet Institute for Global Climate and Ecology.

On the basis of the information available, a synthesized map of ground contamination outside the premises of the combine was set up (Fig. 6.5). Using a method developed at the data processing center of SPA "Typhoon", the data of the ground investigations as well as the map of the gamma dose rates prepared by the Rosgidromet Institute for Global Climate and Ecology were also taken into account. For the estimation of the maximum possible contaminations, a map of the gamma dose rates was set up taking into consideration the upper 90% of the confidence interval (Fig. 6.6).

As is shown by the map of ground contamination outside the premises of the combine (Fig. 6.7), the contaminated area within the radioactive trace having a gamma dose exceeding 15 μ R/h is 89 km² in size. The total energy released of the radioactive products in this area amounts to 3420 μ R× km²/h. According to the map taking into consideration the upper 90% of the confidence interval (Fig. 6.8, upper estimate), these values amount to 135 km² and 7150 μ R × km²/h, respectively.

By means of the values obtained by analysis of the 11 snow samples, the following mean radionuclide composition of the trace was determined: Ru-106 - 31%, Ru-103 - 1%, Nb-95 - 45%, Zr-95 - 22% (Annex 1). In precipitation samples, additional trace amounts of Pu-239 and Sr-90 were found. For the determination of the contamination density, the following dose rates were obtained at 1 m height and a contamination of 1 Ci/km² (with a surface film contamination being assumed): Ru-106 - 3.4 μ R/h, Ru-103 - 8.2 μ R/h, Nb-95 - 13 μ R/h and Zr-95 - 12 μ R/h. With the help of the ratios obtained, maps of the contamination densities of the major radionuclides were prepared (Figs. 6.9 through 6.11). The total of the radioactive products in the area covered by the trace is 350 (730) Ci. For the individual radionuclides, the following values are obtained: Ru-106 - 105 (220) Ci, Ru-103 - 5 (10) Ci, Nb-95 - 165 (345) Ci, Zr-95 - 75 (155) Ci. The estimates taking into account 90% of the confidence interval are given in brackets. To estimate the radionuclide concentration released, a model developed by the data processing center of SPA "Typhoon" was applied for the reproduction of the source parameters. On the basis of the information processed, solution of the inverse problem led to the following estimates of the radionuclide content released: Ru-106 - 300 Ci, Ru-103 - 10 Ci, Nb-95 - 470 Ci and Zr-95 - 210 Ci. Hence, a total activity of 990 Ci was released. For the estimation of the Pu-239 content released (0.2 Ci), the Zr-95/Pu-239 activity ratio of 1000 determined by sample analysis was used.

The ground contamination densities are represented in Figs. 6.12 through 6.15 for the most important radionuclides. They were obtained by simulation taking into account the estimated radionuclide concentrations released. For these calculations, a stochastic model was applied. It was also developed by the data processing center of SPA "Typhoon". Simulation was accomplished for two sources (building affected (height of the source up to 30 m) and ventilation stack (height of the source up to 200 m), respectively) assuming that 50% of the total activity was released by the lower source and that radioactive fallout from the cloud amounted to 0.2 m/s.

Variation of the Pu-239 concentration in the cloud at an altitude of 1 m on the road from Tomsk to Samus and in the village of Georgievka is shown in Fig. 6.16 and Fig. 6.17, respectively. The concentrations of the most important radionuclides calculated as a function of time and space by simulation allowed the individual effective dose equivalent to be estimated for the population of Georgievka when passing the radioactive cloud (Fig. 6.18). The integral dose for 1 hour is about 1 mrem. Here, the share of Pu-239 in the dose due to inhalation is 0.5 mrem. The dose values were calculated in accordance with the method described in [15].



Figure 6.7.



Figure 6.8.

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Figure 6.9. TOMSK – Map of the gamma dose rates as determined by the aerial surveys.

Risø-R-750(EN)



Figure 6.10. TOMSK - Map of the Ru-106 contamination density according to the values measured (Ci/km^2) .

Riso-R-750(EN)



Figure 6.11. TOMSK - Map of the Nb-95 contamination density according to the values measured (Ci/km^2) .



Figure 6.12. TOMSK - Map of the Zr-95 contamination density according to the values measured (Ci/km^2) .

Riso-R-750(EN)



Figure 6.13. TOMSK ~ Map of the Ru-106 contamination density according to the simulation results of SPA "TYPHOON" (Ci/km²).



Figure 6.14. TOMSK - Map of the Nb-95 contamination density according to the simulation results of SPA "TYPHOON" (Ci/km²).

Risø-R-750(EN)



Figure 6.15. TOMSK - Map of the Zr-95 contamination density according to the simulation results of SPA "TYPHOON" (Ci/km²).



Figure 6.16. TOMSK - Map of the Pu-239 contamination density according to the simulation results of SPA "TYPHOON" (Ci/km²).



Figure 6.17.

Riso-R-750(EN)



Figure 6.18. TOMSK - Individual effective dose equivalent according to the simulation results of SPA "Typhoon" (Sv).

 $Ris \phi - R - 750(EN)$

Concluding Remarks

- 1. On the basis of the work (ground investigations and gamma aerial surveys) carried out jointly by the Rosgidromet organizations and Berezovgeologiya, data on the radiation exposure in Russia were obtained shortly after the accident of April 6, 1993 already. These data were transmitted to interested institutions as well as to local and central public information authorities.
- 2. The measurements performed on April 11 and 12, 1993 indicated that within the isolines of 10 μ R/h a contaminated area of up to 25 km in length and up to 6 km in width extended towards the northeastern direction. Thus, the contaminated area outside of the premises of the combine covered about 100 km².
- 3. The total amount of radioactive substances in this area was 530 590 Ci. Isotope composition of the radioactive trace was determined by ruthenium-103 (1%), ruthenium-106 (31%), zirconium-95 (22%), niobium-95 (45%) and plutonium-239 (0.02%).
- 4. A typical feature of the contamination is its considerable heterogeneity. It is caused by the existence of "hot" particles with an activity of up to 10-11 Ci/particle.
- 5. The small village of Georgievka is situated in the contaminated area. Here, gamma exposure rate varied between 14 and 42 μ R/h at 1 m height. Conservative estimation of the maximum external radiation dose of the population of Georgievka yielded a value of 100 mrem/year.
- 6. The Pu-239 inhalation dose of the population of Georgievka when passing the radioactive cloud did not exceed 1.5 mrem.
- 7. A prognosis was made with regard to water contamination of the rivers Samuska and Tom during the flood in spring. Furthermore, contamination of the air layer adjacent to the ground resulting from the wind transport of radionuclides due to agricultural work or the passing of vehicles in the summer months at Georgievka was predicted. The values were far below the limits fixed in accordance with the valid radiation protection regulations.

At the same time, it must be pointed out, however, that radionuclide concentration of the snow water may exceed the limits specified for drinking water. Water contamination should therefore be measured in wells or small closed natural waters.

8. According to the data measured by the meteorological stations, the radioactive products were not entrained beyond the borders of the country.

Annex 1

Calculation of the Factors of Conversion of the Dose Rate Distribution of the Ground into the Contamination Density of Individual Radionuclides

On the basis of the gamma aerial survey of the contaminated area, a map was set up illustrating the distribution of the gamma exposure rate of the ground. For the preparation of maps of ground contamination by individual radionuclides, the factors of conversion of the total dose rate N_{γ} into the surface contamination density of each *i*-th radiosiotope A_i must be known:

$$A_i = B_i N_\gamma \tag{1}$$

Dose rate of the ground is determined by adding the gamma radiations of the individual isotopes

$$N_{\gamma} = \sum_{i}^{n} K_{i} A_{i} \quad (n = 1, 2, ..., n),$$
⁽²⁾

where K_i is the dose coefficient in $(\mu R/h)/(Ci/km^2)$. With the activity share of each *i*-th isotope in the total contamination density A being $p_i = A_i/A$, equation (2) may also be expressed as follows:

$$N_{\gamma} = A \sum_{i}^{n} K_{i} p_{i} = A K .$$
(3)

Here, $K = \sum_{i} K_{i} p_{i}$ is related to the actual mixture of n isotopes investigated. From this, the total ground contamination density by all isotopes may be derived:

$$A = \left(\sum_{i}^{n} K_{i} p_{i}\right)^{-1} N_{\gamma} = B N_{\gamma} .$$
(4)

As $A_i = p_i A$, the following formula for the calculation of the ground contamination density by the *i*-th isotope is obtained from (4):

$$A_i = \frac{p_i}{\sum\limits_{i}^{n} K_i p_i} N_{\gamma} .$$
(5)

Hence, the factors of conversion of the gamma dose rate into the surface contamination density can be represented as follows:

$$V = \left(\sum_{i}^{n} K_{i} p_{i}\right)^{-1}; \quad B_{i} = p_{i} \left(\sum_{i}^{n} K_{i} p_{i}\right)^{-1}.$$
 (6)

Obviously, the conversion factors B and B_i are dependent on the isotope composition of the gamma emitters and their dose factors only.

Let us now determine the formula for the conversion of the gamma exposure rate of the ground measured $N_{0\gamma}$ into the density of isotope contamination of the surface A_i for the radioactive trace generated following the accident at the chemical combine on April 6, 1993.

Sampling of the snow and simultaneous measurements of the dose rate in the contaminated area were performed by ZapSibgidromet experts along the routes M1 - M4 both away from and near the road from Tomsk to Samus (up to 50 m) as well as in the most contaminated section at km 28. As the snow near the road had already begun to thaw, samples were also taken from the upper soil layer.

All samples were observed to have stable isotopic ratios. The activities of the individual isotopes in percent are given below.

Isotope	¹⁰³ Ru	¹⁰⁶ Ru	95Zr	⁹⁵ Nb
i	1	2	3	4
Mean value for routes M1-M4	1.5	3 2.4	22.8	43.3
Mean value for km 28	1.3	29.8	20.8	48.1
Mean value	1.4	31.1	21.8	45.7

For the calculation of the dose coefficients K_i on the basis of the experimental data, equation system (2) with four unknown quantities K_1, K_2, K_3 and K_4 must be solved. Unfortunately, the measurements of A_i near the road turned out to be too small, which obviously had to be attributed to partial penetration of the isotopes into the soil. Correct sampling of the ground was very difficult due to the partly molten snow. Snow samples taken in the taiga far away from the road indicated that practically the entire activity was concentrated on the upper 1 -2 cm of the snow layer [23]. Due to the very small number of measured values available and the strong fluctuations of the results for A_i , which were caused by the existence of "hot" particles in the samples, ⁹⁵Zr and ⁹⁵Nb could only be approximated, $K_3 \approx K_4 = 17(\mu R/h)/(Ci/km^2)$. This nearly corresponds to the theoretical value of K = 14 which was determined when calculating the radiation exposure after the Chernobyl accident [24]. For ¹⁰³Ru and ¹⁰⁶Ru, estimations of this type could not be performed on the basis of the data measured in the Tomsk trace, as the values of A_1 and K_2 were too small. For this reason, the theoretical values of Ki for film contamination of an infinite, homogeneously contaminated plane indicated in [15] were applied in the following calculations:

Isotope	¹⁰³ Ru	¹⁰⁶ Ru	95Zr	⁹⁵ Nb
$K_i(\mu R/h)/(Ci/km^2)$	8.2	3.4	12	13

Using these values of K_i and the above values of p_i , a total of the radioactive products of the Tomsk accident of $K = 9.7(\mu R/h)/(Ci/km^2)$ is obtained. This corresponds to a value of B = 0.115 (Ci/km²)/($\mu R/h$).

According to (1) and (6), ground contamination density of the individual isotopes can be calculated as follows:

For ¹⁰³Ru
$$A_1 = 1.4 \cdot 10^{-3} (N_{\gamma 0} - N_{\gamma \phi})$$
 . (7)

For
106
Ru $A_2 = 3.2 \cdot 10^{-2} (N_{\gamma 0} - N_{\gamma d})$ (8)

For
95
Zr $A_3 = 2.2 \cdot 10^{-2} (N_{\gamma 0} - N_{\gamma \phi})$. (9)

For ⁹⁵Nb
$$A_4 = 4.7 \cdot 10^{-2} (N_{\gamma 0} - N_{\gamma \phi})$$
 (10)

For the total of these isotopes

$$A = 0.115(N_{\gamma 0} - N_{\gamma \phi}) \quad , \tag{11}$$

where $N_{\gamma 0}$ is the dose rate measured and $N_{\gamma \phi}$ the gamma background in $\mu R/h$. The density of radioactive contamination of the ground is denoted by A_i or A in Ci/km².

In accordance with the isotopic ratio 239,240 Pu/ 95 Zr = 3.4 ×10⁻⁴, the formula for the estimation of the contamination density of the ground by these isotopes has the form of

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$$A_{pu} = 7.6 \cdot 10^{-6} (N_{\gamma 0} - N_{\gamma \phi}) \quad \text{Ci/km}^2 \quad . \tag{12}$$

In March 1993 already, an aerial survey of the gamma background had been carried out by ZapSibgidromet along several routes. On the basis of the values obtained, a gamma background of $N_{\gamma\phi} \approx 10\mu$ R/h can be selected for the formulas (7) through (12).

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Analysis and prognosis of radiation exposure following the accident at the Siberian chemical combine Tomsk-7

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