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RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT IN THE AREAS OF LOCATION OF OBJECTS OF THE NUCLEAR FUEL CYCLE

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Abstract

The entry of radionuclides into the environment occurs at all stages of the Nuclear Fuel Cycle: during the extraction and reprocessing of uranium ore, production of fuel elements, operation of nuclear reactors, reprocessing of spent fuel, and burial of radioactive wastes. This report shows the generalization of data in the areas of location of objects of the nuclear fuel cycle (NFC) in the territory of Russia.

INTRODUCTION

Generalized data on the quantities and radionuclide composition of releases to the atmosphere from nuclear facilities of the Ministry of Atomic Energy (Minatom) of Russia are presented in Tables I and II [1, 2].

Among natural radionuclides, ^{222}Rn is a major contributor to the activity of atmospheric releases (about 420 TBq/year). Among artificial radionuclides, isotopes of inert radioactive gases (IRG) have the greatest activity in atmospheric releases of enterprises of Minatom (about 28 PBq). Releases of tritium amount to 410 TBq/year. An appreciable contribution to the IRG activity is made by gas-aerosol releases from NPPs. At the present time, 29 power units with a total installed capacity of 21.242 GWt (el.) are functioning in Russia. Table III presents normalized estimates of radioactive releases from NPPs in Russia in the period 1985-1993. In all, gas-aerosol releases from NPPs with the RBMK-type reactors are distinctly higher than those with the WWER-type reactors. There is a tendency for a decrease in the activity of atmospheric releases in the period 1991-1993, as compared to the preceding period 1985-1990, which is associated with the improvement of the gas purification system at NPPs.

Table IV presents the radionuclide composition of liquid discharges from enterprises of Minatom in 1992-1993 to the surface waters [1, 2]. In 1992, the total activity of liquid discharges amounted to 11.5 PBq. A major contribution to this activity was made by short-lived radionuclides (^{24}Na, ^{31}Si and ^{64}Cu) with a half-life less than 24 hrs, which accounted for about 81%, and by radionuclides with a half-life from 24 hrs to 1 month (^{32}P, ^{51}Cr, ^{56}Mn, ^{76}As, ^{239}Np and others), which accounted for about 18%. The activity of long-lived nuclides with a halflife over 1 year amounted to 9 TBq or less than 1% of the total activity of discharges. A major contribution to the activity of long-lived nuclides was made by tritium, ^{60}Co, ^{106}Ru, ^{90}Sr, ^{137}Cs and others. In 1993, a considerable decrease was observed in the activity of radioactive discharges to the surface waters, as compared to 1992. In 1993, the total activity of liquid wastes amounted to about 1.2 PBq, i.e. decreased by an order of magnitude, as compared to 1992. This decrease in the activity of radionuclide discharges was due to the reduced output at the enterprises of Minatom and removal from service of a number of nuclear facilities.

The bulk of radionuclides enters with liquid discharges from the enterprises of Minatom into the rivers of the Arctic basin (the Yenisei, Tom' and Techa Rivers) [1, 2].

TABLE I **Releases of radionulcides into the atmosphere from the enterprises of Minatom in 1992-1993 [1,2], Bq/year**

Composition of releases	1992 r.	1993 r.
Inert radioactive gases	3.0 E(16)	1.5 E(16)
Isotopes of iodine	1.6 E(11)	7.7 E(10)
Tritium	3.8 E(14)	3.9 E(14)
Isotopes of U, Pu and transuranic elements (natural and artificial)	8.5 E(10)	8.2 E(10)
Rn-222	3.6 E(14)	4.7 E(14)

Below are considered the characteristics of radioactive contamination of the environment during the operation of objects of NFC involved in the extraction of uranium ore and its enrichment, as well as the production of fuel elements, and reprocessing of irradiated nuclear fuel and radioactive wastes.

Argun' River Regional Mining and Chemical Production Association (MCPA)

This MCPA is located in Krasnokamensk district of Chita Region 400 km to the south-east of the city of Chita. It extracts uranium ore from open-cut and underground mines, and enriches it. The sources of radioactive contamination of the environment are the following: an open-cut pit raising dust, terraces of open-cut mines, a tail repository , and a hydrometallurgical plant on uranium ore enrichment. This MCPA also extracts coal and molybdenum ores, which results in chemical contamination of the environment [1, 2].

Mine waters from the Argun' River Regional MCPA are discharged to the surface storing reservoirs and water bodies of the open hydrographic system. Data on the amounts of radionuclides discharged in 1993 are given in Table V. Table VI presents some estimates of the radiation situation paramenters in the zone of observations around the MCPA in 1993. The estimates presented indicate that the soil and vegetation radioactivity is mainly determined by natural radionuclide ^{40}K. The distribution of the total activity of beta emitters in water and atmospheric fallout samples within the area under observation is relatively uniform.

Production Association "Novosibirsk Plant of Chemical Concentrates" (PA "Chemconcentrate")

This PA is located in the northern part of the city of Novosibirsk [1]. It reprocessed uranium concentrates, and now it produces fuel elements for nuclear power engineering. A tail repository for burial of radioactive wastes containing radionuclides of the uranium series, lithium and mercury is located at a distance of 4 km from the PA. Radioactive wastes are delivered to the repository as sludge through the sludge pipeline. Before this pipeline was put into service, wastes were delivered by motor transport, and the environment was contaminated along the route of the delivery.

There are several potential sources of radioactive contamination of the environment at the PA "Chemconcentrate": atmospheric releases of natural and enriched uranium through ventilation systems; the entry of ^{222}Rn into the atmosphere from the surface layer of buried wastes; and the entry of radionuclides into the environment during accidents and the sludge pipeline failure.

TABLE II **The radionuclide composition of the atmospheric releases from the enterprises of Minatom in 1991-1993 [1,2], Bq/year**

Composition of releases	1991	1992	1993	Average
A. Natural radionuclides				
Enriched uranium	1.5 E(9)	1.2 E(9)	1.3 E(9)	1.3 E(9)
Natural uranium	1.0 E(9)	6.6 E(10)	6.1 E(10)	4.3 E(10)
U-238	2.8 E(10)	1.4 E(10)	1.4 E(10)	1.9 E(10)
Other alpha emitters	3.2 E(9)	7.8 E(8)	3.1 E(9)	2.4 E(9)
Rn-222	–	3.6 E(14)	4.7 E(14)	4.2 E(14)
B. Artificial radionuclides				
Na-24	4.4 E(8)	1.9 E(9)	–	1.2 E(9)
P-32	2.8 E(10)	5.2 E(10)	4.8 E(10)	4.3 E(10)
Sc-46	2.2 E(8)	1.9 E(8)	1.9 E(8)	2.0 E(8)
Cr-51	6.3 E(10)	6.8 E(10)	4.1 E(10)	5.7 E(10)
Mn-54, 56	9.6 E(8)	2.0 E(9)	1.9 E(9)	1.6 E(9)
Fe-55, 59	5.6 E(8)	5.6 E(8)	5.6 E(8)	5.6 E(8)
Co-57, 58, 60	2.3 E(9)	4.0 E(9)	4.1 E(9)	3.5 E(9)
Zn-65	1.2 E(9)	9.3 E(8)	8.9 E(8)	1.0 E(9)
Rb-88, 89	9.6 E(10)	7.6 E(10)	–	8.6 E(10)
Sr-89, 90	4.4 E(9)	5.7 E(9)	3.6 E(9)	4.6 E(9)
Zr-95+N_B-95	2.6 E(10)	2.4 E(10)	1.6 E(10)	2.2 E(10)
Mo-99+Tc-99	1.1 E(8)	3.7 E(9)	7.0 E(8)	1.5 E(9)
Ru-103, 106	2.3 E(10)	2.1 E(10)	2.3 E(10)	2.2 E(10)
Sb-124, 125	4.3 E(9)	1.1 E(8)	1.1 E(8)	1.5 E(9)
Cs-134, 137, 138	1.4 E(11)	1.1 E(11)	2.2 E(10)	9.1 E(10)
Ce-141,144	1.7 E(10)	1.6 E(10)	1.2 E(10)	1.5 E(10)
Ba-139, 140	7.3 E(9)	4.7 E(9)	1.5 E(10)	9.0 E(9)
La-140	–	3.7 E(7)	–	3.7 E(7)
Bi-214	1.9 E(10)	3.1 E(9)	–	1.1 E(10)
Pb-212, 214	4.1 E(9)	2.2 E(8)	–	2.2 E(9)
H-3	4.5 E(14)	3.9 E(14)	3.9 E(14)	4.1 E(14)
Isotpoes of I	1.2 E(11)	1.6 E(11)	7.7 E(10)	1.2 E(11)
Total activity of alpha emitters	4.3 E(9)	3.7 E(9)	3.0 E(9)	3.7 E(9)
Isotopes of Pu	3.7 E(8)	3.3 E(8)	3.3 E(8)	3.4 E(8)
Inert radioactive gases, including:	3.8 E(16)	3.0 E(16)	1.5 E(16)	2.8 E(16)
Ar-41	1.6 E(16)	1.3 E(16)	1.1 E(15)	1.0 E(16)
Long-lived nuclides	1.1 E(11)	1.1 E(11)	4.4 E(10)	8.8 E(10)
Short-lived nuclides	–	1.7 E(12)	2.8 E(11)	1.0 E(12)

TABLE III Radioactive releases from NPPs of Russia in the period 1985-1993 [1,5], GBq/GWt(el.)/year

Type of Reactor	Years	Inert radioactive gases	Long-lived nuclides	I-131
WWER	1985-1990	(2.2±0.8) E(5)	2.8±0.2	1.3±0.8
	1991-1993	(5.6±4.0) E(4)	1.7	1.0
RBMK	1985-1990	(2.0±0.4) E(6)	11±7	8.0±3.0
	1991-1993	(1.7±0.9) E(5)	4.6±1.9	2.8±1.4

Note: NPPs with the WWER type reactors are Balakov, Kalinin, Kola and Novovoronezh ; NPPs with the RBMK type reactors are Kursk, Leningrad and Smolensk.

Table VII presents some parameters for the assessment of the radiation situation in the zone of observations around the PA "Chemconcentrate" in 1992. According to observational data, the total activity of beta-emitting nuclides in the atmospheric fallout and the air was within the regional radiation background variations [1]. The exposure dose rate (EDR) in the 100-km zone of observations was 0.07-0.11 μGy/hr on the average, i.e. was also within the natural background variations. At the same time, anomalous patches were detected in the Kalinin district of the city of Novosibirsk, with levels of EDR as high as 2.1μGy/hr around the PA "Chemconcentrate". A probable cause for the formation of these patches is the violation of regulations of solid radioactive waste transportation. Gamma radiation exposure dose rates in the area of location of the sludge pipeline were basically within the range from the background values to 0.35 μGy/hr. A spectrometric analysis of soil samples collected in the area of location of the sludge pipeline indicated that they contained ^{226}Ra and ^{232}Th in amounts exceeding the background values. In the 5-km zone around the tail repository, the values of EDR varied from the background ones to 0.44μGy/hr. In the north-eastern part of the tail repository, a patch with an area of about 3 m^2 and a gamma radiation level approximately 9 μGy/hr was detected.

The activities of beta emitters in potable ground waters are characterized by high variability and vary from 11 to 440 Bq/l. At the present time, it is difficult to make any unambiguous inference about contamination of the ground waters around the tail repository for lack of the system of radioactive contamination monitoring of ground waters.

Siberian Chemical Industrial Complex (SCIC)

This industrial complex is located in the town of Tomsk-7 on the right bank of the Tom' River 15 km to the north of the city of Tomsk. It is the largest compex on the production of plutonium, uranium and transuranic elements. The SCIC comprises the following plants which are the sources of radioactive and chemical contamination of the environment:

- a reactor plant with commercial uranium-graphite reactors for the production of weapon-grade plutonium and electric energy;
- a plant on isotope separation for the production of enriched uranium hexafluoride;
- a plant on production of uranium protoxide-monoxide and uranium hexafluoride;
- a radiochemical plant on reprocessing of irradiated standard lumps to obtain and treat uranium and plutonium salts;
- chemical-metallurgical works.

TABLE IV **Discharges of radionuclides into the surface waters from the enterprises of Minatom in 1992-1993, Bq/year**

Radionuclide	1992	1993
H-3	4.0 E(12)	3.3 E(12)
Na-24	7.5 E(15)	9.5 E(14)
Si-31	1.6 E(15)	2.4 E(13)
P-32	2.2 E(14)	4.6 E(13)
Sc-46	1.7 E(12)	1.3 E(11)
Cr-51	1.3 E(14)	8.3 E(12)
Mn-54	1.3 E(12)	1.7 E(10)
Mn-56	1.5 E(15)	9.0 E(13)
Fe-59	3.2 E(12)	5.1 E(10)
Co-58	9.4 E(12)	7.9 E(10)
Co-60	2.9 E(12)	1.1 E(11)
Cu-64	2.2 E(14)	1.1 E(13)
Zn-65	2.4 E(12)	7.0 E(10)
As-76	1.2 E(14)	3.0 E(12)
Sr-90	6.1 E(11)	1.1 E(12)
Zr-95	5.0 E(11)	4.9 E(10)
Nb-95	3.4 E(11)	5.2 E(10)
Ru-103	3.4 E(11)	1.0 E(10)
Ru-106	1.3 E(12)	2.6 E(10)
I-131	2.2 E(12)	6.1 E(10)
Cs-134	1.9 E(9)	2.0 E(9)
Cs-137	2.2 E(11)	6.3 E(10)
Ba-140	1.7 E(12)	5.2 E(10)
Ce-141	6.1 E(11)	1.7 E(10)
Ce-144	7.1 E(11)	1.0 E(11)
Pb-210	3.0 E(9)	2.1 E(9)
Po-210	1.2 E(10)	1.1 E(10)
Ra-226	5.4 E(9)	4.1 E(9)
Np-239	1.7 E(14)	8.7 E(12)
Pu-239	1.6 E(8)	–
Total activity including:	1.154 E(16)	1.16 E(15)
a) Short-lived nuclides (T ½ < 24 hrs)	9.4 E(15)	9.85 E(14)
b) Radionuclides with 24hrs < T ½ <30 days	2.1 E(15)	1.7 E(14)
c) Long-lived nuclides (T ½ > 1 month)	3.0 E(13)	5.2 E(12)
d) Long-lived nuclides (T ½ > 1 year)	9.0 E(12)	4.6 E(12)

TABLE V **Discharges of radionuclides with sewage waters from the Argun' River Regional MCPA in 1993, Bq/year**

Radionuclide	Total	Natural water bodies	Technological storage ponds
^{240}Pb	1.6 E(10)	2.1 E(9)	1.4 E(10)
^{210}Po	8.0 E(9)	1.6 E(9)	6.4 E(9)
^{226}Ra	1.8 E(10)	2.7 E(9)	1.5 E(10)
Th	7.4 E(9)	2.5 E(9)	4.9 E(9)
U	9.9 E(9)	3.5 E(9)	9.6 E(11)

TABLE VI **Parameters for the radiation situation assessment in the zone of observations around the Argun' River Regional MCPA in 1993**

Parameter (object) of assessment	Dimension	Average value	Maximum value
1. Exposure dose rate	μGy/hr	0.11±0.02(14)	0.14
2. Specific activity of radionuclides			
Soil	Bq/kg		
^{40}K		820±190 (14)	1100
^{137}Cs		40±26 (6)	80
^{226}Ra		60±9 (14)	78
^{232}Th		29±4 (11)	35
Vegetation	Bq/kg		
^{40}K		950±510 (11)	1900
Water			
Total activity of beta emitters	Bq/l	1.6±1.2 (14)	4.4
Atmospheric fallout (total activity of beta emitters)	Bq/m^2 per day	3.0±0.9 (60)	7.2

Note: The number of measurements is indicated in paranthesis. A radius of the zone of observations on the activity of the atmospheric fallout is 100 km and on the other parameters 20 km.

TABLE VII Some parameters for the radiation situation assessment in the zone of observations around the Novosibirsk PA "Chemconcentrate" in 1992

Parameter of assessment	Dimension	Average value	Maximum value
Exposure dose rate	μGy/hr	0.09±0.02 (11000)	2.1
Average monthly activity of beta emitters in the atmospheric fallout	Bq/m^2 per day	1.0±0.7 (24)	9.4
Average monthly activity of beta emitters in the atmospheric air	μBq/m^3	90±23 (12)	180
Total activity of beta emitters in ground waters	Bq/1	100±90 (9)	440

Note: The number of measurements is indicated in parenthesis. A radius of the area of observations on the exposure dose rate is 100 km and on the activity of ground waters 30 km

TABLE VIII Releases and discharges of radionuclides from the Siberian Chemical Industrial Complex in 1992 [1,7] Bq/year

Composition of releases (discharges)	Activity
Releases into the atmosphere	
Inert radioactive gases	7.6 E(15)
^{90}Sr	2.3 E(9)
^{131}I	1.6 E(10)
Total activity of alpha emitters ($^{234, 235, 238}$U, ^{239}P, ^{241}Am)	3.0 E(9)
Discharges to the Tom' River	
^{32}P	4.7 E(13)
^{24}Na	7.1 E(14)
^{46}Sc	1.0 E(11)
^{51}Cr	2.0 E(12)
^{60}Co	1.8 E(10)
^{239}Np	5.8 E(12)

The commercial channel-type reactors with graphite moderators were commissioned in 1958-1963. Three of them were shut down in 1990-1992, and the remaining two should operate till 1995. Since the start-up of the reactors, weapons-grade plutonium was produced, and fuel elements and spent nuclear fuel were reprocessed [6, 7].

Table VIII presents the quantities of radioactive releases and discharges from the SCIC in 1992. As a rule, the atmospheric releases were only fractions of a percent of the maximum permissible releases (MPR). However, for the water discharges of ecologically significant nuclides, such as ^{24}Na and ^{32}P, the discharged activity values were close to the maximum permissible discharges (MPD) or slightly exceeded them. An excess over MPD for ^{32}P in 1992 was mainly due to the lack of installations for desalinization of water supplied for nuclear reactor cooling. Putting into operation of such an installation in 1993 made it possible to decrease appreciably the activity of the water discharges of ^{32}P.

TABLE IX Parameters for the radiation situation assessment in the zone of observations around the Siberian Chemical Industrial complex in 1992

Parameter of assessment	Dimension	Average value	Maximum value
Exposure dose rate	μGy/hr	0.10±0.01	0.12
Average monthly activity of beta emitters in the atmospheric fallout	Bq/m^2 per day	0.74±0.28 (109)	1.8
Soil (in the Tom' River floodplain)	Bq/kg		
^{51}Cr		80±60 (4)	465
^{54}Mn		18±9 (13)	65
^{59}Fe		12±5 (2)	78
^{60}Co		53±26 (12)	265
^{65}Zn		190±100 (12)	950
^{137}Cs		43±30 (18)	640
^{141}Ce		14±6 (7)	23
^{144}Ce		54±26 (2)	80
^{152}Eu		9 (1)	–
Total beta emitters		400±260 (23)	1460
Exposure dose rate	μGy/hr	0.16±0.10 (24)	0.75
Grass	Bq/kg		
^{54}Mn		68 (1)	–
^{60}Co		39 (1)	–
^{65}Zn		680 (1)	–
^{137}Cs		100+60 (2)	160
Needles	Bq/kg		
^{54}Mn		120 (1)	–
^{65}Zn		640±360 (3)	890
^{137}Cs		76 (1)	–
Total beta emitters in the Tom' River water	Bq/l	2.0±1.2 (23)	3300
Ground Waters	$Bq.m^3$		
(wells of the Kantes plot)		36±5 (2)	41
^{90}Sr		22±7 (2)	29

Note: The number of measurements is indicated in parenthesis.

The SCIC is also the source of chemical contamination of the environment. In 1992, discharges of hazardous chemicals (in % of MPD) amounted to 2.4 for fluorides, 1.7 for ammonia, 0.5 for nitrogen oxides, 0.2 for nitric acid, 0.016 for tributyl phosphate, 0.008 for paraffins, 0.001 for carbon tetrachloride, 0.008 for acetone, and 0.001 for benzene [7].

Fifty repositories of liquid and solid radioactive wastes (RW) are located in the territory of the SCIC. In open storage facilities, 4.7 E(18) Bq of RW have been accumulated, and in underground strata, 1.5 E(19) Bq of liquid RW have been pumped at a depth up to 300 m [7, 8].

Table IX presents some parameters of the radiation situation in the zone of observations around the SCIC in 1992. The values of EDR activity of beta emitters in the atmospheric fallout in the 100-km zone around the SCIC do not differ significantly from the regional radiation background. At the same time, radioactive contamination of water and soil is observed in some parts of the floodplain of the Romashka and Tom' Rivers where radioactive discharges from the SCIC have entered for a long time. The presence of ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{141}Ce, ^{144}Ce and ^{152}En was detected in soil samples collected in the Tom' River floodplain. Technogenic radionuclides ^{54}Mn, ^{60}Co and ^{65}Zn which are typical of discharges from the SCIC were detected in samples of vegetation (grass and needles). Technogenic radionuclides were also found in ground waters in amounts no greater than the permissible levels.

On April 6, 1993 at 12.58 local time, an accident with the release of radionuclides into the environment occurred at the radiochemical plant of the SCIC.

The total area of contamination with levels of EDR higher than a radiation background of 0.09 μGy/hr (10 μr/hr) was estimated in April 1993 at about 100 km^2 [2]. The dominant radionuclides in snow samples from the contaminated area were ^{95}Zr, ^{95}Nb, ^{106}Ru and ^{103}Ru. The traces of ^{239}Pu and ^{144}Ce were detected as well. A nonuniform structure of the field of radioactive contamination of the area determined by the presence of hot particles in the composition of radioactive products of the accident deposited onto the snow was revealed [2]. According to observational data, radioactive contamination of the environment with the products of the accident at the SCIC was of a local nature. The accident resulted in the formation of a narrow radioactive trace 35-45 km long in a north-eastern direction from the SCIC (from the trace concentrations of ^{95}Zr and ^{95}Nb in soil). There are no populated places in the territory of the trace, except for the village of Georgievka. The external exposure dose to inhabitants of Georgievka from the products of the accident over 50 years of permanent residence will amount to 0.22-0.31 mSv which is negligible, as compared to the dose from the natural radiation background. On a whole, the radiation accident at the SCIC has not led to significant radiological consequences for the population [2, 7].

In the process of gamma survey of the area around the SCIC, sites contaminated with ^{137}Cs up to 37-74 kBq/m^2 were detected. They fan out within 10 km of the SCIC to the north, 2-3 km to the west and east, as well as along the shore line of-the Tom' River. The nature of this contamination does not allow relating it to the accident occurred in April 1993. Most likely, this contamination is the result of the SCIC operation of many years [2].

In conclusion, it should be pointed out that it is essential to develop an integrated radioecological monitoring of the radionuclide content in the atmosphere, soil, surface and ground waters, natural and agricultural food chains. Data of the radioecological monitoring are necessary for further analysis of risks posed by radioactive and chemical contamination of the environment, and for the development,of immediate measures to insure ecological safety during the operation of enterprises of NFC in Russia.

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