CHRONOLOGICAL ANALYSIS OF CONTAMINATION OF THE RUSSIAN TERRITORY BY LONG-LIVED RADIONUCLIDES

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ABSTRACT

Contamination with biologically significant long-lived ^{137}Cs and ^{90}Sr radionuclides is observed in the entire territory of the Russian Federation. The sources of radioactive contamination of the dry land territory were surface, underground and atmospheric tests of nuclear weapons, underground peaceful nuclear explosions, regular and accidental releases from nuclear industry objects and nuclear power plants, accidents with military equipment. The spatial picture of the contamination was formed under the influence of processes of global, long-range, intermediate and local atmospheric transfer of radionuclides from the sources of contamination. Further evolution of contamination occurred as a result of radioactive decay and, to a lesser extent, processes of soil and atmospheric migration of radionuclides. A chronological analysis of published data allowed us to distinguish the main stages of contamination from 1949 to the present time.

Keywords: *contamination, radionuclides, Russia, land, periods*

INTRODUCTION

The entire territory of the Russian Federation was subjected to contamination by artificial long-lived radionuclides to one degree or another at various times. A large number of scientific publications are devoted to the study of radioactive contamination all over the world, including that in Russia.

Spatial and temporal characteristics of contamination, as well as their isotopic composition, are determined by the sources and character of radioactive releases, meteorological conditions of atmospheric transport and deposition on the earth's surface, the processes of soil, wind, aquatic and biological migration, and by radioactive decay processes. Atmospheric fallout of radionuclides may be local, remote (in the process of tropospheric transport from a source of contamination), and global, formed by deposition of fine radioactive particles from the stratosphere.

The goal of this paper is to analyze and establish chronological categorization of the dry land contamination in Russia with biologically significant long-lived radionuclides ^{137}Cs and ^{90}Sr based on the previously published scientific data.

PROCESSES AND CATEGORIZATION

1949-1953. First Nuclear Tests. Low-Yield Explosions

On August 29, 1949, having conducted the first nuclear explosion with an energy release of about 20 kilotons in TNT equivalent at a test site located 170 km from the city of Semipalatinsk, the USSR began nuclear weapons tests. The first nuclear explosions were low-yield and led to local and remote radioactive fallout patterns. A spatial distribution of individual radioactive contamination patterns depended on the parameters of an explosion (yield and height of the explosion), as well as on meteorological conditions in the area of the passage of the radioactive cloud. During this period, nuclear tests were conducted at the Semipalatinsk test site in the former Soviet Republic of Kazakhstan. These tests led to serious radioactive contamination of both the test site itself and the adjacent territories. To be more specific, in Russia were affected the Altai region (explosions of August 29, 1949, August 7, 1962, January 15, 1965). The Republic of Tuva, the Krasnoyarsk Region, the Omsk, Tomsk, Novosibirsk, Kemerovo and Irkutsk regions were polluted to a lesser extent.

In 1991 during an airborne gamma-spectral survey only isolated spots of increased ^{137}Cs contamination of soil (from 0.1 to 0.3 Ci/km^2) were detected in the south-west of the Altai Region. These fallouts may be associated with nuclear testing 29.08. 1949 or with ^{137}Cs global fallout. In this case, it is almost impossible to separate ^{137}Cs from a specific explosion and ^{137}Cs due to global fallout [1].

The start of nuclear weapon testing was preceded by the launch of nuclear industry enterprises. Their activities are accompanied by routine release of radionuclides into the atmosphere, which also create contamination of the underlying surface.

During this period, radioactive contamination affected the soils in the floodplains of Techa and Iset rivers as a result of the discharge of liquid radioactive waste from "Mayak" plant reactors into the open hydrographic network. For the period from 1949 to 1956 76 million m^3 of liquid radioactive waste with a total activity of 2.75 million Ci was discharged. Currently, the main dose-forming radionuclides are ^{137}Cs and ^{90}Sr. Soil contamination in the floodplain of the rivers was formed as a result of regular (lower tier of the floodplain) or occasional (upper tier of the floodplain) flooding. ^{90}Sr is highly soluble in water and migrates downstream, resulting in ^{90}Sr contamination found throughout the floodplain of the Techa and Iset rivers. ^{137}Cs is less mobile, hence high levels of ^{137}Cs are concentrated mainly in the floodplain of the Techa River. In the head water of the river Techa contamination levels of ^{90}Sr could reach more than 100 Ci/km^2, and ^{137}Cs - 500 Ci/km^2. Downstream, anomalously high contamination values (up to 200-300 Ci/km^2 for ^{137}Cs and up to 150 Ci/km^2 for ^{90}Sr) were related to swampy floodplain areas. In the Iset river floodplain, the largest pollution was observed on the 15-kilometer distance below the Techa estuary and was 4–16 Ci/km^2 for ^{90}Sr and 1–2 Ci/km^2 for ^{137}Cs [2].

1953-1963. Atmospheric Thermonuclear Weapon Tests. High-Yield Explosions. Global Atmospheric Fallouts

The global and intense radioactive contamination of the Earth began with the launching of atmospheric testing of high-yield thermonuclear weapons. In 1951, tests of thermonuclear weapons began in the United States, and on August 12, 1953 a thermonuclear device with an energy release of 400 kt was detonated at the Soviet Semipalatinsk test site. Global fallout was formed from highly dispersed particles (size of particle was a one or a fraction of micron) [3], released into the stratosphere during explosions. A distinguishing feature of global fallout is the long-term (up to several years) presence of products from different nuclear explosions in the stratosphere. The main isotopes of global fallout are long-lived ^{90}Sr, ^{137}Cs and to a lesser extent ^{3}H. The maximum intensity of global fallout was observed at the beginning of the 1960s. According to the UN Scientific Committee since the start of nuclear testing until 1963, 19.3 MCi ^{90}Sr and 33 MCi ^{137}Cs were released into the atmosphere [4].

The main studies of the global contamination by ^{90}Sr and ^{137}Cs were carried out in the middle 60s - early 70s. Detailed information on the distribution of global ^{137}Cs obtained using airborne gamma-spectral surveys is available for the territory of the former USSR. The average deposition density of ^{137}Cs in soils on the USSR territory was 92 mCi/km^2 (ranging from 15 to 200 mCi/km^2) in 1974 [5]. The spatial distribution of the ^{137}Cs deposition in soils was distinctly related to the annual precipitation level. Contamination levels increased when approaching mountainous areas, which may also be associated with an increase in the average annual rainfall here. Global fallout had a clear latitudinal zonality. Maximum levels of contamination were located at the latitudes of 50 - 60º. To the south and north contamination levels declined.

The ^{90}Sr and ^{137}Cs contamination of global origin became widespread all over the Earth. It caused great concern and led to the signing of the Moscow Treaty Banning Nuclear Weapon Tests in the Atmosphere, in Outer Space and Under Water in 1963 [3]. Since then only France and China continued nuclear tests in the atmosphere until 1974 and 1980 respectively.

In this period of time, tests of nuclear weapons continued at the Semipalatinsk test site. On September 21, 1955, tests at the Novaya Zemlya test site began. Tests in the atmosphere of the most high-yield thermonuclear bombs had been carried out at this site until the end of 1962. Remote radioactive traces resulting from a series of tests in 1961 were discovered on a vast territory of Russia to the east and west of the Urals [3]. Due to the specifics of high-yield air explosions, the intensity of tropospheric ^{137}Cs and ^{90}Sr fallout on the soil was low, but this series of explosions led to a marked increase in global fallout. The only ground-based test of a nuclear weapon carried out at the Novaya Zemlya test site on September 7, 1957, in addition to contaminating a part of the territory of the test site itself, led to atmospheric deposition in the adjacent part of the continental territory of Russia.

On September 14, 1954, in the course maneuvers, an atomic bomb with a yield of 40 kilotons was exploded at the Totsk military training area in the Orenburg region at the altitude of 350 meters above the ground. According to the data obtained in 1996-1999 the presence of ^{152}Eu and ^{60}Co isotopes was revealed at the epicenter of the explosion. Within the local trace, there was a slight increase in the content of ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ in the soil compared with the average global fallout parameters typical for the Orenburg region. At a distance of 50 - 70 km from the epicenter, there maximum soil contamination. On the whole, the territory of the local trace of the Totsk airborne nuclear explosion is currently radiation-safe. [6].

On September 29, 1957, a chemical explosion of a storage facility for radioactive waste occurred at the “Mayak” plant at the Urals region. As a result of the explosion, radionuclides were released into the environment with a total activity of about 20 MCi Main part of the emission settled down within the industrial site, and 2 MCi spread in the northeastern direction, forming the East-Ural Radioactive Trail. The composition of the release is the following: ^{90}Sr + ^{90}Y - 5.4%, ^{95}Zr + ^{95}Nb - 24.9%, ^{144}Ce + ^{144}Pr - 66%, ^{106}Ru + ^{106}Rh - 3.7%, ^{137}Cs- 0.039% [3]. Currently, the main dose-forming radionuclide in this area is ^{90}Sr. Maps of contamination could be found in the Atlas [2] which was developed basing on the results of 1995–2012 field studies.

For many years the main sources of technology-related radionuclides entering the Yenisei river has been two reactors of the Krasnoyarsk nuclear plant. It is located 40 km from Krasnoyarsk downstream the Yenisei River. The first of them was launched in August 1958, the second - in 1961. Both reactors were phased out in 1992. The study [7] showed that the soil contamination with ^{137}Cs of the Yenisei valley propagated to the inflow of the river into the Arctic Ocean. Soil contamination levels with ^{137}Cs reached 4 Ci/km^2, and such contamination levels could be observed at large distances from the source of contamination (480-800 km). Contamination levels at the estuary of the river could reach 0.5 Ci/km^2.

1963-1986. Underground Nuclear Tests and Peaceful Nuclear Explosions. Individual Accidents

After signing in 1963 of the Moscow Treaty, only underground tests of nuclear weapons were conducted at the Semipalatinsk and Novaya Zemlya test sites. Underground tests at the Semipalatinsk test site which is located outside of territory of Russia did not contribute to its radioactive contamination. Underground explosions at the Novaya Zemlya test site in some cases led to local contamination of the test site.

In addition, underground nuclear explosions for industrial and scientific purposes were conducted on the territory of the Russian Federation. A total of 81 explosions were carried out. The explosions were carried out for the following purposes: deep seismic sounding of the earth's crust; liquidation of emergency gas fountains; the creation of underground storage facilities; disposal of toxic industrial effluents; intensification of oil and gas production; crushing ore; gas abatement in coal mines; and others [8]. More than half of the technological sites where underground nuclear explosions were carried out have been closed and the radiation levels on the surface have regional background values. Facilities for extraction of oil, disposal of industrial effluents and underground tanks for gas condensate storage are in operation. The process of their operation is under constant radiation monitoring.

There is a small group of objects, the surface near which is contaminated with radioactive products. These are incomplete camouflet explosions: "Kraton-3" (Sakha Republic (Yakutia), 1978) and “Globus-1"(Ivanovo Region, 1971), as well as explosions with a release of soil: "Taiga" (northern Perm Region, 1971) and Kristall (Sakha Republic (Yakutia), 1974). Near these objects there are local areas with increased levels of radiation and soil contamination with ^{137}Cs, ^{90}Sr, ^{60}Co, $^{239+240}$Pu and other radionuclides.

In 1967, wind transportation of radioactive dust from dried-up Lake Karachay became another source of radioactive contamination near “Mayak” plant in the Chelyabinsk region. Lake Karachay had served as the collector of liquid radioactive waste from the plant. The wind carried away about 0.6 MCi of radioactivity. The contamination area was estimated at 2,700 km^2 outside the territory of the industrial zone [2].

The contamination caused by the accident on August 10, 1985 on the nuclear submarine K-431 at the ship repair plant in Chazhma bay near Vladivostok, mainly affected the sea area of the Ussuriysk bay. On the Danube Peninsula, adjacent to the plant, the contamination density of ^{137}Cs and ^{90}Sr is estimated to be 0.07-0.001 Ci/km^2. The main part of the activity (90-99%) on the trail at the time of the fallout occurred at ^{60}Co [9].

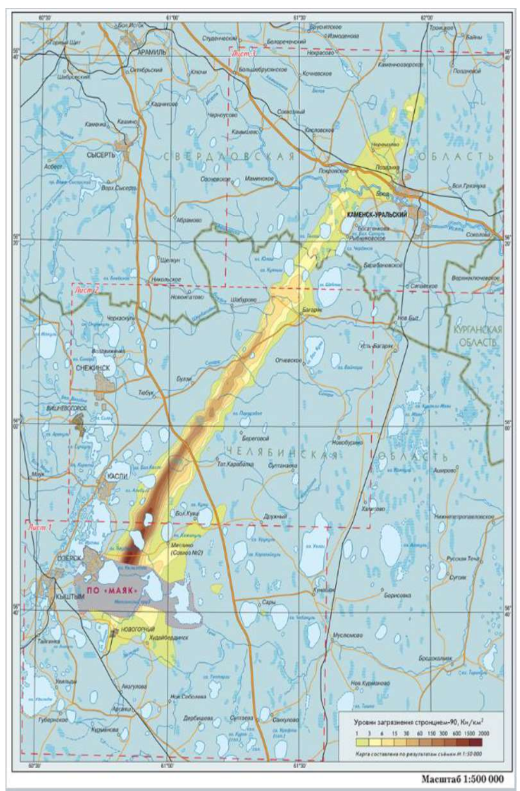

Figure 1 ^{90}Sr contamination in the “Mayak” plant area of influence [2]

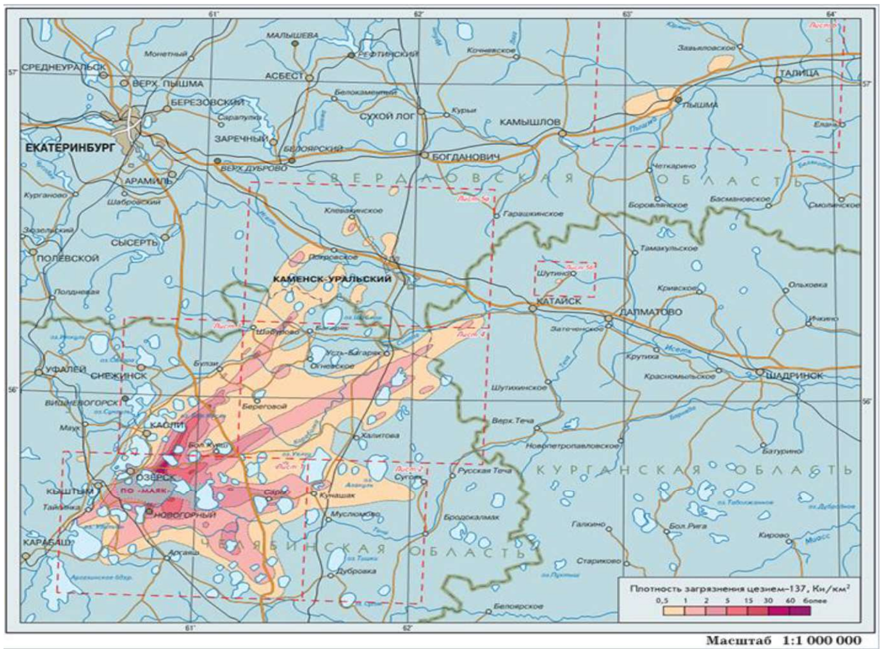

Figure 2 ^{137}Cs contamination in the "Mayak" plant area of influence [2]

1986. Chernobyl Nuclear Power Plant Accident

The accident of April 26, 1986 at the fourth power unit of the Chernobyl NPP resulted in destruction of the reactor, reactor block and machine room and release into the environment of radionuclides, which far exceeded all radionuclides releases from previous accidents of atomic reactors. Two thermal explosions caused the initial short-term release of radionuclides into the environment. This was followed by the fire of reactor's graphite masonry supported by the decay energy of radionuclides inside the reactor. The result of these processes was continuous release to the atmosphere a powerful jet of gaseous and aerosol radioactive products. The liquidation of the fire resulted in a significant reduction in the release of radioactivity into the atmosphere. According to [10], the total release of radioactivity from the reactor (with the exception of inert gases) by May 6, 1986 was about 50 MCi or 3.5% of the total activity accumulated in the reactor.

Radioactive contamination after the Chernobyl accident affected vast territories. In addition to Belarus, Ukraine and Russia, significant contamination was observed in many European countries [12]. Contamination of areas with a level exceeding 0.2 Ci/km^2 is almost entirely due to the Chernobyl accident. The area of ^{137}Cs contamination exceeding 0.2 Ci/km^2 on the European territory of Russia was 535.02 thousand km^2. Levels above 1 Ci/km^2 were observed in 19 regions of Russia and in December 1993 their area was 57.65 thousand km^2 [13].

The structure of radionuclide fallout on the Earth's surface was determined by the long-term (11 days) radionuclide release from the damaged reactor and continuously changing meteorological parameters: speed and direction of wind transfer, intensity of radionuclide leaching from the atmosphere by precipitation.

In 1988 - 1993 a large-scale study was carried out on the territory of the European part of the former USSR, the Urals and Western Siberia using the airborne gamma-spectral survey method combined with soil sampling and analysis. As a result of this work, maps of ^{137}Cs (which was the main dose-forming long-lived Chernobyl radionuclide) soil contamination of territories were developed. These maps were published in Atlases [11], [12]. The map of ^{137}Cs soil contamination in Russia in 1986 is presented in Figure 3.

Figure 3 Map of ^{137}Cs contamination of Russia by the end of May 1986 (Ci/km^2) [2]

1987 – Present. Post-Chernobyl Small Accidents

During this period there were no events resulting in significant radioactive contamination of territories. Underground nuclear explosions, the last of which was carried out at the Novaya Zemlya test site on October 24, 1990, were not accompanied by releases of radionuclides into the environment. By the mid-1980s, global stratospheric fallout of ^{137}Cs and ^{90}Sr had practically ceased.

There were no significant accidents at nuclear power plants and enterprises of the nuclear industry. The exception is the accident of April 6, 1993 at the Siberian Chemical Combine (Tomsk-7) which caused local contamination. The total amount of radioactive products deposited outside the industrial zone was estimated at 530-590 Ci. These products were mainly radionuclides with relatively short half-live times (up to 1 year). The area of contamination, estimated by the authors on the base of published data [14], amounted to 90 km^2.

Another exception was the radiation accident of 2013 at a heavy engineering plant in the city of Elektrostal (Moscow region). The resulting ^{137}Cs contamination exceeded the background level at the latitude of Moscow by 18 to 750 times [15].

However, it had the character of small area spots. Accident at the Japanese nuclear power plant “Fukushima - 1” on March 11, 2011, did not lead to noticeable soil contamination in Russia [15].

CONCLUSION

Radioactive contamination with ^{137}Cs and ^{90}Sr on the dry land territory of Russia was formed as a result of many events of different nature that took place at different times since 1949. The field of contamination has a complex spatially heterogeneous character.

Chronological analysis allowed identifying 5 main stages of contamination: 1949-1953 (from the start of operation of the nuclear industry and the first Soviet nuclear test to the beginning of global radioactive fallout); 1953-1963 (until the end of the Soviet and American nuclear tests in the atmosphere); 1963-1986 (period between the Moscow Treaty Banning Nuclear Weapon Tests in the Atmosphere, in Outer Space and Under Water and the Chernobyl accident); 1986 (the accident at the Chernobyl Nuclear Power Plant) and finally the period after Chernobyl, which continues to the present day.

At present, in the absence of major radiation accidents and other events, the density of contamination decreases at a rate approximately equal to the rate of radioactive decay, i.e. decreases twice in about 30 years for both ^{137}Cs and ^{90}Sr.

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