

**A REVIEW OF DATA ON RELEASES OF RADIOACTIVE WASTES
FROM THE “MAYAK” PRODUCTION ASSOCIATION
INTO THE TECHA RIVER IN 1949-1956**

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1. INTRODUCTION

Operation of the Mayak Production Association (Rosatom, Ozersk, Chelyabinsk region) for production of weapon plutonium in 1940-1950s resulted in significant radioactive contamination of the environment, including water bodies of the Urals Region.

Releases of the liquid radioactive wastes into the Techa River in 1949-1956 resulted in intensive contamination of the river that remains till nowadays. Switching of the main releases of the liquid radioactive wastes from the Techa River to Karachay Lake, a decrease in the activity of the released solutions and creation of the cascades of storage reservoirs in the upper Techa-region (Techa River cascades, TRC) allowed improvement in radiation situation. However, radioactive contamination of the Techa River bottom sediments and floodplain and continuous entrance of the radionuclides through bypass channels (with the seepage from TRC reservoirs and other sources) has continuous negative impact on the situation that does not comply with the current sanitary-ecological guidelines.

Long-term medical observations of the populations in the Techa River villages exposed as a result of radioactive releases and radioecological monitoring in the environment allowed accumulation of large amounts of radioecological, medical and dosimetric data. Radiation and social protection of the exposed populations and estimation of radiation risks from chronic exposure require reliable evaluation of the radiation situation and estimation of doses of the internal and external exposure. To implement these tasks, specialists from four Russian organizations (Urals Research Center for Radiation Medicine, “Mayak” Production Association, Russian Federal Nuclear Center – Zababakhin Institute of Technical Physics and Institute of Plants & Animals Ecology of Urals Division of Russian Academy of Sciences) and international colleagues established Project #2841 in 2005-2007 devoted to the reconstruction of the releases from “Mayak” PA into the Techa River in 1949-1956 under the International Science and Technology Center.

The Project allowed detailed analysis of a number of archive documents that were not available previously because of secrecy (Ratner et al. 1950; Perminov et al. 1950; Alexandrov et al. 1951; Demyanovich et al. 1952a,b; Ilyin 1956). This resulted in more precise estimation of the total activity and radionuclide composition of the releases into the Techa River in 1949-1956. Data analysis, presented in this report, is substantially based on data provided by Mayak PA specialists in their technical report on ISTC Project 2841 (Glagolenko et al. 2006a).

2. DESCRIPTION OF THE MAYAK PRODUCTION ASSOCIATION AS THE SOURCE OF THE TECHA RIVER CONTAMINATION

The Mayak Production Association (MPA) was created in the Southern Urals at the end of the 1940s for the production of weapon plutonium and reprocessing of fission products (FP). In 1948-1949 it comprised one uranium-graphite reactor at nominal 100-MW power, operating with thermal neutrons and using direct flow water-cooling loops (Facility A); a radiochemical plant for the extraction of ^{239}Pu from uranium irradiated in the reactors (Facility B, which included the radioactive waste-storage facilities known as Complex C); and a chemical-metallurgical plant for the production and machining of metallic plutonium (Facility V). An extensive increase in plutonium production and a simultaneous adjustment of the technology of its production occurred during the first period of the MPA operation. Radiochemical processing (in Facility B) resulted in large volumes of liquid radioactive wastes (LRW) of different specific activities. The absence of reliable waste-management and storage technologies resulted in significant radioactive contamination of the Techa River and other water bodies located near the MPA during 1949-1956 and exposure of the population living along the river.

Releases of radioactive wastes into the Techa River commenced in January 1949. Systematic control of the discharges was started only in September 1951. Radioactive wastes were released from different stages of technological reprocessing of irradiated uranium blocks, therefore, they differed by the age, radionuclide composition and physical-chemical properties that determined their behavior in the river system. For retrospective evaluation of the total activity and radionuclide composition of uranium-fission-product mixture discharged into the Techa River in 1949-1956, it is necessary to trace the technological chain of reprocessing of radioactive material from unloading irradiated uranium blocks from reactor to entrance of FP into the Techa River. Special attention should be paid to the period from January 1950 to October 1951 because from November 1951 the main releases were routed to closed Karachay Lake.

2.1. The reactor plant (Facility A)

June 19, 1948 is considered as the start of work of Facility A when the first industrial uranium-graphite reactor was put into operation at nominal 100-MW power (so called "reactor A"). According to technology, average duration of irradiation of uranium blocks in the reactor should be about 120 days (Glagolenko et al. 2006b). It is known, that the rate of plutonium accumulation in uranium blocks depends on the power distribution along the radius and the height of the active zone of a reactor. Since the specific heat power at the central part of the active zone was greater than at the peripheral part, different technological channels required irradiation for different times in order to obtain necessary amounts of plutonium. Several months after the launch, the reactor was switched to the regime of regular (planned) refill: about 1% of uranium blocks were unloaded and downloaded into the active zone of the reactor each day (Glagolenko et al. 2006b). Reactor design allowed unloading and downloading of uranium blocks in any technological channel while the reactor was in operation.

The reactor was cooled with the waters of Kyzyl-Tash Lake, in which Techa River takes its beginning and floods through dam D-2. Since 1948, hot waters after cooling of the reactors were released into Kyzyl-Tash Lake. Neutron irradiation resulted in formation of radioactive isotopes in the cooling waters flooding through the reactor (induced radioactivity). Besides this, cooling pools and storage shafts, where unloaded from the reactor irradiated uranium blocks were stored for decay of ^{239}Np , were filled with waters from Kyzyl-Tash Lake. Since aluminium cladding of some uranium blocks was damaged, waters discharged from Facility A into Kyzyl-Tash Lake contained some amounts of uranium-fission isotopes. Therefore, the Techa River water at the exit from

Kyzyl-Tash Lake was contaminated with radionuclides since 1948; however, releases from Facility A were negligible compared to the releases from radiochemical plant.

Storage of irradiated uranium blocks in cooling pools of Facility A allowed significant reduction in total activity of uranium fission products accumulated in the uranium blocks during their irradiation in the reactors due to radioactive decay of short-lived radionuclides. Because uranium FP are released during radiochemical processing, the hold-up time of irradiated uranium blocks in cooling pools of Facility A is a very important parameter for evaluation of radionuclide composition of the releases into the Techa River and, therefore, for the assessment of doses for exposed population.

It was possible to find in MPA archives monthly information on the mass balance of irradiated uranium in Facility A for the period from December 1948 to 1954 (Glagolenko et al. 2006b). This allowed evaluation of monthly-averaged values of effective hold-up time of uranium blocks conveyed from cooling pools of Facility A to radiochemical reprocessing (Fig. 1).

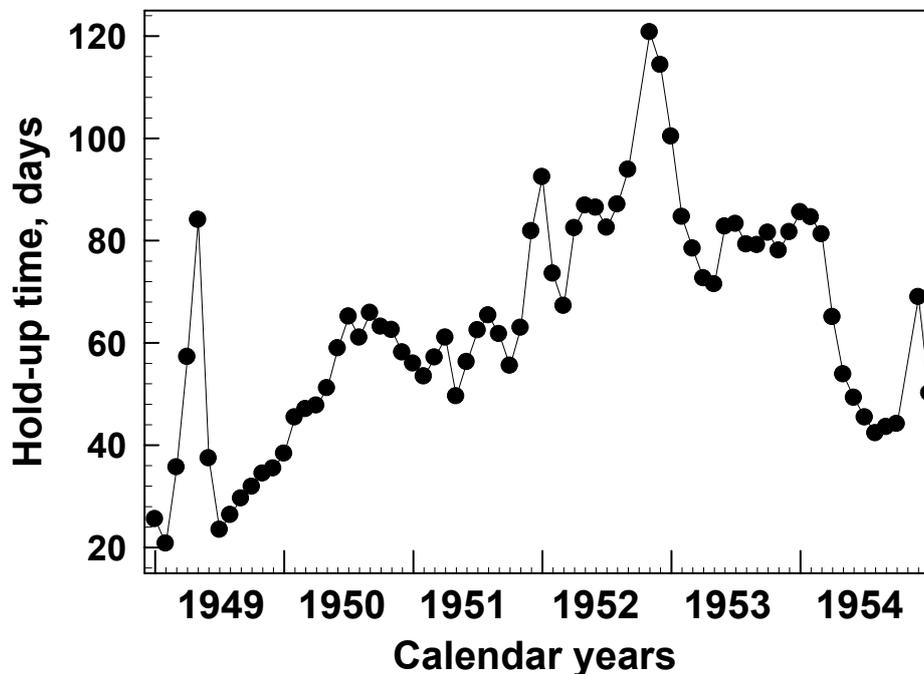


Fig. 1. Monthly-averaged effective hold-up time of irradiated uranium blocks conveyed from Facility A to radiochemical reprocessing in 1949-1954 (according to data of Glagolenko et al. 2006b).

As can be seen from Fig 1, monthly-averaged values of the hold-up time during the first years of MPA operation were in the range from 20 days (January 1949) to 120 days (October 1952). It should be noted that monthly-averaged values of the hold-up time in the period of massive releases into the Techa River (March 1950 – October 1951) varied from 48 to 66 days and the average value for the period of interest is 59 ± 5 days. Therefore, the age of uranium fission products, which determines the ratio between short-lived and long-lived radionuclides in the main releases into the Techa River, could not be less than two months.

The second reactor, AV-1 (300 MW), was put into operation at the MPA in the beginning of April 1950 (Brokhovich 1996, pp. 78 and 95). Since Summer 1950, this reactor worked almost continuously. The first reactor continued working at power higher than the nominal power (Brokhovich 1996, p. 130). The first part of uranium blocks irradiated in reactor AV-1 was unloaded in August 1950 and conveyed to radiochemical plant in September 1950 (Tryakin 1998). Since this time, irradiated uranium blocks from both reactors were reprocessed in Facility B. The third reactor AV-2 was put into operation in April 1951; however, irradiated uranium blocks from this reactor were conveyed to radiochemical plant in December 1951 after the releases of liquid radioactive wastes had been switched from the Techa River to closed Karachay Lake.

2.2. The radiochemical plant (Facility B)

December 22, 1948 is considered as the date of beginning of the operation of Facility B (the radiochemical plant where significant amounts of LRW originated) when the first portion of irradiated uranium blocks was conveyed from Facility A to Facility B. It should be noted, that irradiated uranium, conveyed from Facility A, was directly sent to reprocessing. For this reason, the amount of irradiated uranium reflects the capacity of Facility B. Since the amount of the main technological LRW is proportional to the amount of reprocessed uranium, the dynamics in Facility B output is extremely important for evaluation of the total activity of discharges into the Techa River.

Analysis of data on monthly mass balance of irradiated uranium conveyed from Facility A to Facility B in 1950-1951 showed that there was a slight increase in the capacity of Facility B in January-July, 1950. Then, there was a sharp decrease in the number of reprocessed uranium blocks in August 1950 explained by the fact that there was a significant repair in Facility B aimed at the reduction of the number of radiation sources and alteration in the system for release of relief gases into the main stack (Gladyshev 1992, p. 28-29). Since September 1950 the capacity of Facility B increased three-fold compared to average capacity in the first half of the year. This is explained by the fact, that Facility B commenced reprocessing of irradiated uranium blocks from two reactors. Notable reduction in the capacity, observed in October-November 1951, was seemingly related to replacement of technological equipment and regular repair. Sharp increase in the capacity of facility B occurred after December 1951 due to reprocessing of irradiated uranium blocks from three reactors.

It is known that not all types of LRW originated during plutonium production were released into the Techa River. High-level LRW (HLW), containing large amounts of uranium and plutonium, were conveyed for long-term storage to specially equipped tanks. Besides this, during the first stages of MPA operation, some types of LRW were routed from Facility B to a bog named "Staroe Boloto" and to so called "chromate holes". To reproduce the picture of the Techa River contamination it is necessary to generally describe the technological processes occurred in facility B in 1949-1951. Table 1 and Fig. 2 represent the structure and functions of the main production departments of Facility B in 1949-1951.

Table 1. The main production departments of Facility B in 1949–1951.

Department number	Basic technological functions	Comments
Departments 1 and 4	Preparation of chemical reagents	The work in these departments did not deal with radioactivity.
Department 2	Dissolution of aluminum cladding of uranium blocks and of uranium blocks themselves irradiated in the reactor and held in the cooling pools of Facility A	The LRW formed during the dissolution of the cladding was conveyed to Department 13; a solution of irradiated uranium in 60%-nitric acid was conveyed to Department 3. The dissolution of blocks was accompanied by formation of large amounts of radioactive aerosols.
Department 3	Separation of uranium and plutonium from the bulk of the FP by oxidative-acetate precipitation of U and Pu	The precipitate containing U and Pu was solved and conveyed to Department 6. The decantate containing the bulk of FP and residues of U and Pu was conveyed to Department 13. A part of LRW of lower activity was conveyed to Department 16.
Department 6	Separation of plutonium from the bulk of uranium by reductive-acetate precipitation of uranium	The solution of plutonium was conveyed to Department 7; the precipitate (sodium uranyl acetate) was conveyed to Department 15a for extraction of uranium.
Department 7	Alkaline concentration of plutonium (precipitation of Pu and residues of U by NaOH)	The concentrate of plutonium was conveyed to Department 8. The alkaline decantate was conveyed to Department 16.
Department 8	Plutonium extraction using lanthanum-fluoride technology; repeated purification of Pu from residues of FP and U	Pu concentrate was conveyed to Facility V; the decantate with residues of FP, Pu, and U was conveyed to Department 9a.
Department 9a	Long-term settling of the decantate formed in Department 8, containing FP, Pu and U, aimed at the extraction of Pu residues	After long-term settling the precipitate of PuF ₄ was conveyed back to Department 8; a fluoride solution was released into Staroe Boloto.
Department 13	Long-term storage of high-level LRW (HLW) in specially equipped tanks (so called complexes/tanks C) before subsequent reprocessing aimed at extraction of plutonium and uranium residues.	Accidental leaks of HLW into cooling waters and groundwaters occasionally occurred in Department 13. This resulted in entrance of HLW into the Techa River.

Departments 15a and 15	Obtaining of uranium diacetate salt by dissolution of sodium uranyl acetate (from Department 6) in HNO ₃ , manganous purification and precipitation	Department 15 was put into operation in the end of 1950. The solution of sodium uranyl acetate was stored for six months (for decay of ⁹⁵ Zr and ⁹⁵ Nb) before obtaining uranium; LRW as a manganous pulp were conveyed to Department 16.
Department 16	Reprocessing and purification of the LRW from Departments 2, 3, 7, 15a and others by settling and/or filtration in chromate holes; adsorptive purification of LRW.	In 1949, Department 16 received only low-level LRW, which were released into the Techa River after adsorptive purification. Since January 1950, low-level LRW were released into the Techa River without any purification. Medium-level LRW were conveyed to Department 16 since May 1950. They were released into the Techa River after purification, however, a part of LRW were released into the Techa River without any purification.

As follows from the description of the technology used in the radiochemical plant, maximum amounts of LRW of high specific fission-product activity were formed at the first stages of the reprocessing of irradiated uranium blocks: in Department 2 during the dissolution of aluminum cladding of irradiated uranium blocks and of uranium blocks themselves and in Department 3 during the separation of uranium and plutonium from the bulk of fission products with use of an oxidative-acetate precipitation method (Table 1 and Fig. 2). Besides this, significant amounts of technological LRW were formed in subsequent stages of radiochemical processing. The wastes included alkaline decantates formed during the concentrating of Pu solution in Department 7, fluoride solutions from Department 9 (discharged to Staroe Boloto), and solutions of manganous pulp in bisulphite from Department 15a. It is obvious, that the activity of these portions of LRW was lower and the age of FP was greater compared to the age of FP in LRW from Departments 2 and 3. This particularly concerns the solutions of manganous pulp from Department 15a, where the solutions of Pu were stored in a special tank for six months for decay of ⁹⁵Zr and ⁹⁵Nb (Sokhina 2000, p. 29).

It is seen from Fig. 2, that HLW with residues of plutonium and uranium were conveyed to Department 13 for a long-term storage in specially equipped tanks (so called "tanks C") for subsequent reprocessing. Medium-level wastes were conveyed to Department 16 for purification before their discharge into the Techa River. A part of medium-level LRW was held in Department 13 for some time before the purification. LRW were released into the Techa River from collectors A and B through a pumping station. According to the technology, it was supposed that cooling and drainage waters from Department 13 were not in contact with radioactive solutions. However, it was revealed in Autumn 1951, that there were leaks of HLW from tanks C in Department 13.

It should be noted, that Fig. 2 demonstrates only those types of the LRW that mainly determined contamination of the Techa River. It should also be noted that there were continuous technological changes during the first period of the MPA operation that influenced the amount and radionuclide composition of the radioactive releases into the Techa River. Fig. 2 is composed on the basis of a scheme of service lines in Facility B in September 1951 from the report of

Alexandrov et al. (1951). Technological processes related to formation of the main LRW are described in detail in the following sections of the report.

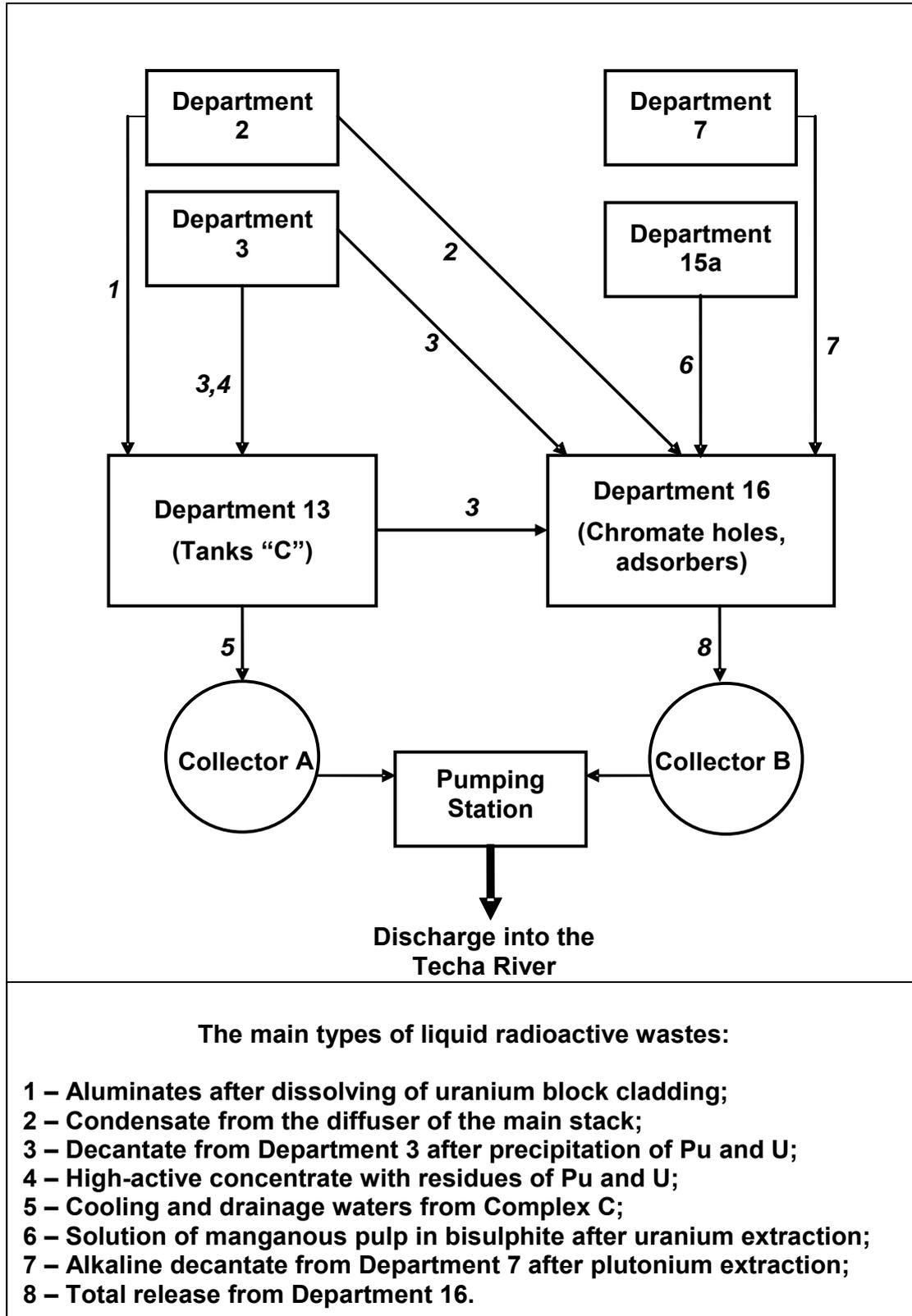


Fig. 2. The main production departments of Facility B where in 1949–1951 the bulk of LRW was formed and the general scheme for conveying of radioactive solutions from the places of their formation to the Techa River

2.2.1. Dissolution of uranium blocks (Department 2 of Facility B)

Dissolution of aluminium cladding of uranium blocks and of uranium blocks themselves irradiated in the reactor and held in the cooling pools of Facility A occurred in Department 2. A solution of irradiated uranium in 60%-nitric acid obtained in Department 2 was conveyed to Department 3. The main types of LRW formed during the dissolution of the cladding were aluminates and condensate from the diffuser of the main stack (Fig. 2).

Solutions of aluminium cladding (so called “product 61”) were delivered to tanks C since Facility B was put into operation (Demyanovich et al. 1952a). Since March 1950, a part of product 61 was preliminary reprocessed in Department 3 resulting in formation of aluminate waters (so called “product 673”) that were discharged into Staroe Boloto (Perminov et al. 1950; Ratner et al. 1950). From September to December 1950, product 673 from Department 3 was conveyed to Department 16 for additional reprocessing and was stored in chromate hole before discharging into the Techa River. Since December 20, 1950, product 673 was released into the Techa River without additional purification (Minutes of the meeting on December 20, 1950).

Dissolution of uranium blocks in Department 2 resulted in formation of significant amounts of radioactive aerosols. Relief gases from technological apparatus (mainly nitric oxides) entered a reinforced concrete pipe, which was a 150-m high and had a diameter on the ground of 11 m and on the top of 6 m. From the beginning of Department 2 operation to Summer 1950, relief gases mixed with ventilating emissions in the pipe, since it was presumed that the draught will be enough to release all gases into the atmosphere (Gladyshev 1992, p. 28-29). However, it was revealed in Summer 1950, that a condensate of vent gases settled on internal walls of the concrete pipe and flowed down into workrooms of Facility B. (Gladyshev 1992, p. 28-29). For this reason, in August 1950, a stainless steel pipe was constructed inside the concrete pipe for relief gases from technological apparatus. The condensate was diverted into special pools. Therefore, since September 1950 the condensate from the diffuser of main stack (CDMS) was collected and conveyed to Department 16 and subsequently discharged into the Techa River. According to D.I. Ilyin (1956), the CDMS was “contaminated by all chemical and radiochemical substances that occurred at the plant”.

Besides this, periodic washing of radioactive contamination from the apparatus and canyons of Department 2 resulted in formation of so called “desorption waters”. Washing of apparatus was conducted when sufficient amount of “deposits” was formed on the walls of apparatus and service lines. These waters were extremely contaminated with chemical substances, because to dissolve “deposits” on the walls of the apparatus, solutions of potassium permanganate, caustic soda, nitric, oxalic, and even hydrofluoric acids were used (Ilyin 1956). Desorption waters were conveyed to Department 13 or Department 16 from where they were discharged into the Techa River. These releases were not planned in the technology design, and their control commenced only in 1952.

2.2.2. Separation of uranium and plutonium from the bulk of fission products (Department 3 of Facility B)

Department 3 of Facility B served for separation of uranium and plutonium from the bulk of FP by oxidative-acetate precipitation of U and Pu. According to L.P. Sokhina (1997) the decantate, in addition to fission products, also contained about 10% of the plutonium; the HLW with large amounts of uranium and plutonium were to be retained for long-term storage in the tanks of Complex C (Department 13 of Facility B). Sokhina writes (2000, p. 41-42),

“The volumes of released decantates and rinsing waters after oxidating-acetate precipitation were enormous. Concentrating by evaporation was considered in the project in order to reduce them, however, the facility for evaporation of the solutions [Ed. Note: Department 5] was not built because specialists of IPCAS [Ed. Note: the Institute of Physical Chemistry of Academy of Sciences] suggested a method of alkaline concentration of stable impurities (iron, chromium, manganese) and of uranium, plutonium and fission products adsorbed on these impurities.”

From the beginning of Facility B operation to the end of November 1949, high-level decantate from Department 3 (so called product 633) was disposed into tanks of storages C-1, C-2 and C-3 (Department 13) without preliminary reprocessing. In the end of November 1949, a new facility was put into operation in Department 3 that allowed separation of LRW into two groups: 1) concentrated wastes containing significant amounts of Pu and U kept for subsequent recycling and 2) middle-level LRW (Alexandrov et al. 1951). The new facility (so called facility 900) allowed obtaining alkaline decantate (“product 901”) with γ -activity of 20-35 mg-eq L⁻¹ and significantly more active concentrate (“product 904”) with accumulated residues of Pu and U (from LRW) from the original decantate using methods suggested by IPCAS. Product 904 was conveyed to Department 13 and product 901 was temporarily discharged into Staroe Boloto from December 1949 to February 17, 1950. After, product 901 was discharged into empty tanks of C-4 (Alexandrov et al. 1951) for temporary storage and conveyed to Department 16 (Fig. 2) for further purification from strontium and chromium using the method of chromate and carbonate precipitation (see Section 2.3).

Considerations of the scheme of acetate precipitation at Facility B (Zilberman 1961, p. 81-82) infer that the decantate from Department 3 contained the bulk of Cs, Sr, Ba, and Ru, whereas the precipitate contained Ce, Y, and Zr along with Pu and U; therefore, the latter was conveyed for subsequent stages of reprocessing. Sokhina also mentions the fact of predominant capture of Zr and Nb by the precipitate formed in Department 3 (Sokhina 2000, p. 27). Therefore, the precipitate containing U and Pu was dissolved and conveyed for subsequent reprocessing at Department 6, where plutonium was separated from the bulk of uranium by reductive-acetate precipitation of uranium.

Besides this, analogous with the situation in Department 2, periodic washing of radioactive contamination from the apparatus and canyons of Department 3 resulted in formation of so called “desorption waters” that were conveyed to Department 13 or Department 16 and further discharged into the Techa River.

2.2.3. Alkaline concentration of plutonium (Department 7 of Facility B)

Alkaline concentration of plutonium was performed in Department 7 (precipitation of Pu and residues of U by NaOH). Plutonium concentrate was conveyed to Department 8 for further reprocessing. The formed alkaline decantate with residues of FP (so called “product 757”) was conveyed to Department 16 (Fig. 2). Product 757 was classified as a low-level product and was discharged into the Techa River since the beginning of Facility B operation.

2.2.4. Obtaining of uranium (Departments 15 and 15a of Facility B)

Departments 15 and 15a served for obtaining of product 80 (uranyl salt) by dissolution of sodium uranyl acetate (from Department 6) in HNO₃, manganous purification and precipitation. The solutions of were stored in special tanks in Department 15a for six months (for decay of ⁹⁵Zr and ⁹⁵Nb) before manganous purification and uranium extraction. Obtaining of uranyl salt

accompanied by formation of so called “product 91” (solution of manganous pulp in bisulphite). Almost 100% of its activity was due to ^{95}Zr and ^{95}Nb (Alexandrov et al. 1951).

Department 15 was put in operation later than 1949. The exact date of its launch could still not be found in MPA archives, however, some indirect data (description of this kind of LRW is absent in the report of Perminov et al. dated July 1950) imply that obtaining of uranium commenced in the end of 1950. Because during the first period of its operation Department 15 was to process the total volume of the solution accumulated by its launch, the daily volume of product 80 and, correspondingly, the amount of LRW (products 91 and 92) at that time did not correlate with the number of irradiated uranium blocks conveyed for reprocessing from Facility A.

According to Alexandrov et al. (1951), in September 1951, products 91 and 92 conveyed to Department 16 from Department 15a, were released into the Techa River without any purification. Description of the technology for reprocessing of LRW of this time could not be found in MPA archives. Taking into consideration that the problem of reprocessing of the pulp of manganese dioxide was not resolved in 1952 (Starik et al. 1952), it can be presumed that this type of LRW was released into the Techa River without any purification since the beginning of uranium production.

2.3. Storage and reprocessing of LRW (Departments 13 and 16 of Facility B)

Storage and reprocessing of LRW in Facility B was performed in Department 13 (which was the storage of high-level radioactive wastes) and in Department 16 (which has “chromate holes” and adsorbers for LRW purification). The history of their operation, described below, is extremely important for the reconstruction of discharges into the Techa River.

2.3.1. High-level liquid radioactive waste storage complex (Complex C)

HLW storage (synonyms are Complex/Building 120, Complexes C, Department 13 of Plant 25/Facility B) was put into operation in the beginning of 1949. Building of new tanks C was ceased in December 1949, when the new facility on reprocessing of high-level wastes commenced operation in Department 3 (Alexandrov et al. 1951). In 1949-1951, decantates from Department 3 and high-level LRW were conveyed into the storage complex. The storage capacity was planned for annual receiving of 15,000 m³ of LRW (Sokhina 2001).

The storage consisted of separate complexes (C-2, C-3 and C-4), representing rectangular canyons embedded in ground (complex C-1 was cooled with air and was not related to discharges into the Techa River, for this reason it is not considered in the report). Inside canyons, there were tanks made of stainless steel; the walls were about 13-mm thick. Each tank was cylindrical with a volume of 250 m³ and was placed in a separate canyon of reinforced concrete (with the inner walls of 1.0-1.5 m thick). The top of the canyon was closed with a cylindrical plate of reinforced concrete; the top of the plate was covered with a layer of soil 1–1.5-m thick. Montejuses were used for transmission and delivery of HLW. The scheme of a storage tank is shown in Fig. 3.

The decay of radionuclides in the tanks was accompanied by considerable energy release (especially during the first period of their filling). This energy release was being compensated for by a water-cooling system. The technological process for cooling the tanks provided continuous water supply underneath into the gap between the walls of a tank and concrete walls of a canyon at the rate of 10-12 m³ per hour (Krasnopeyev and Karpov, 1984). Cooling lines were individual for each particular tank and the water was being supplied alternately to each tank. The outflow lines from canyons had a general collector within the same building. According to the design, all tanks of “Complex C” were placed below the level of groundwater. Because of damaged waterproofing in the canyons of all waste-storage buildings, groundwater flowed into the canyons.

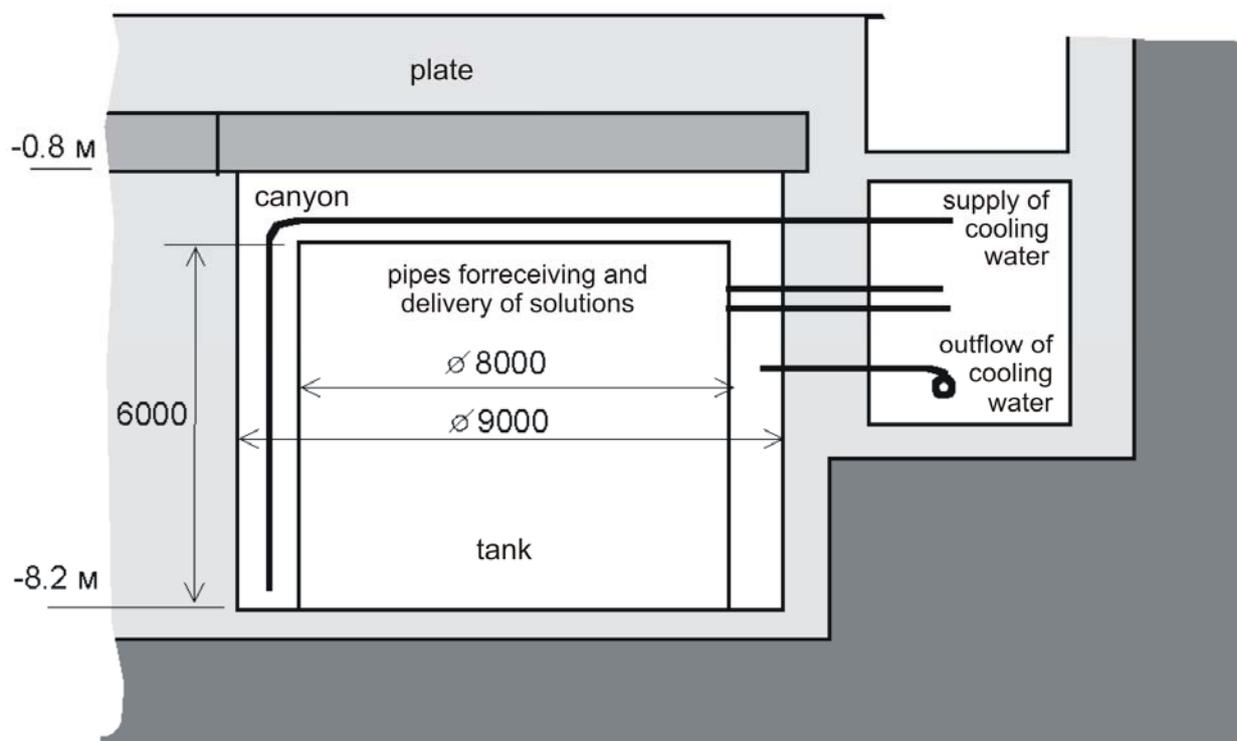


Fig. 3. The design of the tank of Complexes C-2 – C-4 (cross-sectional view).

According to Ilyin (1956), the daily average volume of cooling and drainage waters in Complexes C was $130 \text{ m}^3 \text{ day}^{-1}$. According to data obtained in laboratory of Facility B (Demyanovich et al. 1952a), the daily debit of water in the cooling system of Complex C in 1952 was $15\text{--}20 \text{ m}^3 \text{ day}^{-1}$ and the debit of groundwater discharged from Complex 120 was $15 \text{ m}^3 \text{ day}^{-1}$ (it was noted that the volume of groundwater increased up to $25\text{--}30 \text{ m}^3 \text{ day}^{-1}$ during raining or snow melting). In total, these numbers correspond to $30\text{--}50 \text{ m}^3 \text{ day}^{-1}$, which is lower by a factor of 2.5–3 than estimated by Ilyin (1956). Possible explanation of these differences is that cooling water was delivered to the tanks for several hours a day, at that, large volumes of water were required for cooling of the tanks with “fresh” high-level wastes (HLW), however, cooling of the tanks with “old wastes” was not necessary. Archival documents at Mayak PA contain records that filling of canyons in Complexes 120/2,3,4 C with water became more frequent in October 1950. It was noted that pumping of groundwater out of Complexes C was continuously performed at that time, however, the documents do not contain quantitative estimate of water debit in the cooling system in Complexes C. Cooling water from Complexes C was discharged into the Techa River.

Report by Demyanovich et al. (1952a) contains a figure on filling of tanks C-2 and C-3 in May-December 1949. According to the data, tanks C-2 were filled by products 61 and 633. Until June 30, 1949, the tanks were filled by a mixture of these products (average filling time of one tank with the volume of 250 m^3 was 5 days at that period); since the July, products 61 and 633 were kept separately and the average filling time of a tank by product 633 increased up to 8 days and by product 61 was equal to 3.5 months suggesting that daily debit of product 61 was significantly lower than of product 633. It should also be noted that according to Demyanovich et al. (1952a), content of Pu and U in product 61 was substantially higher than in product 633. Most of tanks C-2 and C-3 were filled up until December 1949 and only two tanks were being filled by product 61 till April 1951 (Demyanovich et al. 1952a). After, according to Demyanovich et al. (1952a), tanks C-2

and C-3 were not filled until 1952 when a possibility for reprocessing of these LRW was considered. It should be noted that the report by Demyanovich et al. (1952a) does not contain information on any damage of tanks in complexes C-2 and C-3, therefore, it could be presumed that LRW were successfully kept till 1952 when their reprocessing commenced.

Complex C-4 that was filled since December 1949 operated in somewhat different regime. First of all, it should be reminded that by that time the new facility (“facility 900”) for LRW reprocessing commenced operation in Department 3 allowing production of high-active concentrate (so called “product 904”) accumulating residues of Pu and U from LRW. Product 904 was conveyed to tanks of complex C-4 for a long-term storage. It is known that the product was classified as HLW that had specific activity of 1 Ci/L or greater. L.P.Sokhina (2000, p. 42), wrote:

“The temperature of a solution in a tank of volume of 250 m³ amounted to 100 degC in 20 days if the specific activity of the solution was 40 Ci/L”.

Thus, to escape boiling of the solution, tanks with HLW were cooled with water to keep the temperature at the level below 50-60°C (Sokhina 2000). It is evident, that specific activity of the solution decreased with time due to radioactive decay of fission products resulting in corresponding decrease in the intensiveness of cooling water supply. Thus, some tanks of C-4 were filled up with HLW that were subjected to subsequent reprocessing aimed at recycling of Pu and U.

Beside this, since February 17, 1950¹ available C-4 tanks were temporarily filled with alkaline decantate from Department 3 (so called “product 901”) with lower specific activity (100-120 mCi/L) and without substantial amounts of Pu and U. According to Perminov et al. (1950), since April 28, 1950, product 901 was conveyed from C-4 tanks to Department 16 for purification with the use of chromate and carbonate precipitation (see Section 2.3.2). In total, 2,800 m³ of product 901 were reprocessed in 2.5 months (i.e. until 15 July 1950 when the report by Perminov et al. (1950) was issued) that allowed to empty 11 tanks in complex C-4. It can be concluded from the Protocol of the meeting of Chief engineer of Facility B on December 20, 1950 (Minutes..., 1950), that a number of C-4 tanks continued operation in the same regime, i.e. they were filled with middle-level product 901 that was later conveyed for reprocessing to Department 16 after 1-3-month storage.

According to the history of Complexes C operation, inflow of groundwater and cooling water into the canyons resulted in floating of storage tanks up from the canyon floors. As a result of floating, the walls and bottoms of the tanks were deformed, which led to flaws in joint welds. Some reports (Karpov et al. 1983; Krasnopeyev and Karpov 1984) indicated that during the first years of complex C-4 operation, 19 tanks of all 20 tanks in the Complex floated. Some of the tanks had damaged or faulty pipes for receiving and delivery of solutions. Because of this damage to the tanks a significant portion of the canyons was filled with a mixture of high-active wastes with water instead of cooling water. This fact was first revealed by the Alexandrov Commission in Autumn 1951 (Alexandrov et al. 1951). Measurements of specific activity of water in canyons in Complex C-4, performed on October 4-9, 1951, revealed significant contamination in several canyons. The report provides the results of measurement of specific activity in canyons of five tanks. Seemingly, they were filled with HLW (product 904)², specific activity of water in canyon of one of these tanks amounted 815 mCi/L. Total activity of water in this canyon was evaluated to be 50,000 Ci on October 9, 1951.

Since the beginning of Complex C operation, the cooling and drainage waters were discharged into the Techa River through emergency reservoir (so called Collector A with a volume

¹ Product 901 was discharged into Staroe Boloto from December 1949 till February 17, 1950.

² For three tanks of five, there is a direct notation in the report that they contained HLW, for the rest two tanks pH of a solution (<7) indicates that they were also filled with HLW.

of 1000 m³) and the pumping station where they mixed with other discharged solutions (Fig. 2). Since the design did not consider radioactive contamination of the cooling waters, dosimetry control in Collector A was absent. According to data from Alexandrov et al. (1951) and Ilyin (1956), cases of unexpectedly high levels of released activity (so called “wild overflows”) into the Techa River observed on September 27-29 and on October 2, 8-11, 26 and 31, 1951. According to estimates of Ilyin (1956), beta-activity of released wastes during the “wild overflows” amounted up to 100,000 Ci day⁻¹.

Since October 28, 1951 releases from Complex 120 were to be temporarily switched to Karachay Lake in case of radioactive contamination in Collector A; only low-level LRW were to be released into the Techa River (Starik et al. 1952). However, “wild overflows” into the Techa River with the activity, amounting up to 20,000 Ci day⁻¹ in particular cases, occurred in the end of 1951 and the first half of 1952 (Ilyin 1956). These releases were recorded by the system of monitoring, implemented directly in the effluent canal exiting from the pumping station into the Techa River (subsection 4.1).

2.3.2. Description of the chromate holes and the history of their operation

The first chromate hole was put into operation on April 28, 1950 (Perminov et al. 1950) for purification of decantate, formed in Department 3, from strontium and chromium by precipitation of barium chromate and barium carbonate.

The hole was located at the distance of 100 m to the north from the building where Department 16 was located. The volume of the hole was 500 m³ and the square was 160 m² (Ratner et al. 1950; Perminov et al. 1950). The chromate hole represented wooden reservoir with the walls made of boards 50-mm thick connected into rabbet. According to the design, the hole was to be waterproof (due to covering of the walls with mineral rubber and making clay locks), however, the design requirements were not completely met when the hole was being built (as a result, the solution delivered into the hole in May and the first half of June completely sank into the ground). The hole was placed in the layer of loamy soil at the depth of 2 meters from the ground level. The thickness of loamy soil at location of the hole was about 6 meters and turned into crumbling porphyryte ground with further layer of solid porphyryte ground. The groundwater at location of the hole was at the level of -7 m deep.

Suspensions of barium chromate and barium carbonate with specific gamma-activity of 10-20 mg-equ L⁻¹ and specific beta-activity of 20–30 mCi L⁻¹ were flowed into the hole. According to the estimates of Perminov et al. (1950), 3,000 m³ of product 922 with the total activity of 30,000-35,000 g-equ Ra had been delivered into the hole by July 15, 1950. About half of the activity associated with precipitate and another half – with solution. From the comparison of these figures with the volume of LRW conveyed from C-4 and reprocessed over the same period (2800 m³) it can be concluded that in Spring-Summer 1950 the first chromate hole mainly reprocessed product 901 from tanks C-4 after its preliminary hold up in Department 13.

The solution in the hole drained through the ground and significantly purified from radioactive dirt adsorbing on the ground. The rate of adsorption was about 180 m³ day⁻¹ in the beginning of May 1950, however, the rate decreased to 36 m³ day⁻¹ by the beginning of June 1950 (Ratner et al. 1950; Perminov et al. 1950). The decrease in filtrating capacity of the hole was explained by plugging of leakages in the inner surface of the hole with barium chromate. The authors wrote,

“Further operation of Department 16 should take into account that the chromate hole will stop filtering the solution, thus the solution conveyed to the hole must be released into the Techa River”.

To prevent releases of the solution into the Techa River, the authors (Perminov et al. 1950) suggested in July 1950 to build additional ground hole without inner covering near the existing hole, so that filtrating solution would flow itself from the old hole into the new hole at the level of 300 m³. Thus, the first hole would serve as a settling tank and the second hole – as a filter.

The authors (Perminov et al. 1950) also evaluated expected contamination of the Techa River for the assessment of the consequences of the release of radioactive solution after its settling in the chromate hole. According to their estimates, daily averaged release of radionuclides into the Techa River was equal to 600 Ci with 99% contribution of ruthenium and cesium into the total activity and less than 1% contribution of strontium in the case of storing of product 922 in chromate hole. Taking into account the dilution of the releases with the river water, the authors presumed that specific activity of radionuclides would not exceed current guidelines for contamination of drinking water (Perminov et al. 1950). The authors indicated that *temporary* release of purified solutions into the Techa River after one-month storage was acceptable; however, long-term *permanent* release was inadmissible due to adsorption of the radionuclides by bottom sediments and fast expansion of the radioactive contamination downstream.

According to archive documents of Facility B, the second hole had not been built by the end of 1950. It is known that technical specifications for the hole' design had been prepared by December 25, 1950, however, it was not successful to find any document at MPA archives that would contain technological parameters of the second chromate hole. It is said in a document dated December 20, 1950 (Minutes 1950), that at that time less-active product 673 (volume 50 m³ day⁻¹; daily release – 50 g-equ Ra) was released into the first hole together with product 901. The volume of these solutions exceeded the designed output of Department 16 (accounted only for reprocessing of product 901) that resulted in insufficient settling of product 922 due to less timing for settling in the hole. It was decided to release unpurified product 673 directly into the Techa River before construction of the second hole. Thus, it can be concluded that product 922 was released into the Techa River in December 1950 after its settling in the first chromate hole. Since December 20, 1950, the amount of daily release increased to due cassation of purification of product 673.

In the beginning of February 1951 the first chromate hole finally stopped filtrating and a “bog”, containing liquid with high-specific activity, formed on the northern side of the chromate hole (Gromov and Gladyshev 1951). The document (Gromov and Gladyshev 1951) says, that:

“Availability of spare tanks in Complexes C allows us to continue working until February 26, 1951, whereupon, the object will be under the evident treat of shutdown if the second chromate hole is not put into operation”.

It is also said that a design for assembling service lines for the second hole (so called “hole for Building 145”) was not completed at the time of preparation of the document, however, the authors provided estimates of expected period of its operation:

“It is supposed that if the same output as during the first months [of 1951] is assumed, the hole of Building 145 will be filled up in August 1951”.

Unfortunately, the date of the beginning of the operation of the second chromate hole could still not be found at MPA archives. It can be supposed that the second filtrating hole was built in Spring 1951. It is also known that in Autumn 1951 operation of the second chromate hole was ceased (Alexandrov et al. 1951):

“Since September 17, the solution after settling in chromate hole, which did not provide filtration due to malfunctions, was released into the Techa River”.

The report by Demyanovich et al. (1952b) provides measurements of specific activity of product 922 performed on October 19-26, 1951, before and after passing the adsorbers. Therefore,

it can be concluded, that release of product 922 from the first chromate hole into the Techa River continued until switching of the main technological releases to Karachay Lake.

It is known, that condensate from the diffuser of the main stack was temporarily released into the first chromate hole in January 1952, and in February 1952 further operation of Department 16 was ceased due to high levels of radioactive contamination (Demyanovich et al. 1952b). As a result, operation of the chromate holes was ceased in the beginning of 1952 and the holes themselves were laid up.

2.3.3. Evaluation of the technological processes for LRW reprocessing in 1949-1951

Analysis of the data described in previous subsections shows that there were two approaches tested for management of the liquid radioactive wastes during the first years of Facility B operation:

1. Long-term storage of high-level LRW with large amounts of uranium and plutonium in closed tanks for radioactive decay of short-lived radionuclides and further reprocessing of the wastes aimed at extraction of uranium and plutonium;
2. Release of middle-level and low-level LRW into open water bodies.

Implementation of these approaches required development of the technology for concentrating of high-level LRW with large amounts of uranium and plutonium into small volumes (for their storage in Facility B). It was necessary to simultaneously develop methods for decreasing the concentration of radionuclides in solutions, released into the environment, down to the levels corresponding to guidelines for open water bodies used by general public for water supply, domestic demands and swimming (Gusev 1956).

According to MPA archive documents, HLW reprocessing aimed at extraction of uranium and plutonium commenced in Facility B in 1952. L.P. Sokhina wrote (2000, p. 41–42),

“The task was to obtain dense precipitates of hydroxide, occupying minimal volumes, and to extract uranium and plutonium from the wastes. Repeated alkaline precipitation of impurities was carried out after the extraction of these valuable elements. Alkaline precipitates were collected, dissolved in nitric acid and conveyed to special storage (Complex C).”

It can be supposed that “wild overflows” commenced in April 1951, when as a result of extraordinary flood, the inflow of groundwater into the canyons Complex C became significant and the storage tanks floated up, which led to flaws in joint welds. It can also be supposed that continuous corrosion increased leaks in the storage tanks; moreover, the releases depended on the seasonal changes in the level of groundwater, which increased in spring due to snow melting and in autumn due to raining. Seemingly, in 1951, the radionuclide composition of the LRW released into the Techa River as a result of accidental leaks of HLW with the cooling and ground waters of Complex C, could correspond to theoretical mixture of FP with the age equal to the sum of the holdup time of uranium blocks in cooling pools of Facility A and the time of storage in the tanks of Complex C.

According to the developed technology, decrease in specific activity of LRW released into the Techa River was achieved by dissolving, adsorption, filtration through ground and settling. The simplest method for decreasing the specific activity of radionuclides in the Techa River was dilution of released wastes with uncontaminated water. This method was used in 1950-1951 by increasing the flow-rate from Kyzyl-Tash Lake into the Techa River through Dam D-2 (Alexandrov et al. 1951; Ilyin 1956). However, application of the method was restricted because large amounts

of water in Kyzyl-Tash Lake were required for cooling of the reactors in Facility A. Moreover, it is evident that application of this method did not result in decrease of the total activity released from Facility B into the environment.

Adsorption allows purification of LRW from activity adsorbed on dredge (fine particles). Until January 1950, all low-level LRW formed in Facility B were passed through adsorbers in Department 16 and released into the Techa River only after their purification (Alexandrov et al. 1951). According to the reports of Ratner et al. (1950) and Perminov et al. (1950), adsorptive purification of the decantates formed in Department 7 with the use of iron ore led to decrease in specific activity by a factor of 8–10. The authors wrote,

“In January [Ed. Note: 1950] after establishing new guidelines for FP release, the adsorptive purification was ceased and solutions formed in Department 7 were directly delivered to the effluent canal bypassing the adsorbers”.

It follows from Alexandrov et al. (1951), that the method of adsorptive purification was more effective for purification of the condensate from the diffuser of the main stack (CDMS) resulting in decrease in specific activity of this type of the LRW by a factor of 100. However, data on the period of adsorptive purification of the CDMS before its release into the Techa River could not be found from available archive documents. This question is extremely important for reliable evaluation of the total activity and radionuclide composition of the releases into the Techa River, since the data, obtained by the laboratory of Facility B in 1952 (Demyanovich et al. 1952b), indicate, that specific activity of the original CDMS was hundreds of mCi L^{-1} , i.e. was comparable and even exceeded specific activity of product 901 (decantate formed in Department 3 and contained the bulk of FP). According to Starik et al. (1952), purification of CDMS at Facility B was absent in 1951. Based on these data, it can be supposed, that release of CDMS in 1950-1951 into the Techa River was one of the factors that determined the levels and radionuclide composition of radioactive contamination of the river water and bottom sediments. It can also be supposed that the radionuclide composition of the CDMS approximately corresponded to the mixture of FP with the age equal to the holdup time of uranium blocks in cooling pools of Facility A.

According to Ratner et al. (1950), in 1950, there were glauconite columns in Department 16 for additional adsorptive purification of product 922 (after its settling in chromate hole). However, the authors (Ratner et al. 1950) indicated low adsorptive capacity of the glauconite and impossibility of using the glauconite columns for long-term operation. The report of Demyanovich et al. (1952b) contains the results of parallel analysis of product 922 before and after the glauconite purification for the period of October 20-24, 1951. These data indicate that adsorptive purification led to decrease in specific activity by a factor of 1.7–1.9.

Chromate holes in Department 16 were used in 1950-1951 for filtering product 922 through the ground and/or for its settling aimed at precipitation of barium chromate and barium carbonate. The history of chromate holes operation, described in subsection 2.3.2, shows that during the period of massive releases into the Techa River only one chromate hole worked and the second chromate hole was put into operation later; the regime of the chromate holes operation changed for several times. In 1949-1951, operation of the chromate holes followed two different regimes:

1. Filtering through the ground (without release into the Techa River);
2. Settling aimed at precipitation and further release of purified solution into the Techa River.

The first chromate hole was filtering for 3-4 months after it had been put into operation in May 1950 (Ratner et al. 1950; Perminov et al. 1950). The second chromate hole operated in the same regime after its launch (Alexandrov et al. 1951). The solution was discharged into the chromate hole, drained into the ground and significantly purified from radioactive impurities

adsorbing on the ground. Studies performed in 1950 indicated, that some amounts of the activity drained from the hole into the groundwater could reach Mishelyak River (a tributary of the Techa River, located at the distance of 7 km from the hole) not earlier than in 20 years. Therefore, it is evident, that when the chromate hole operated at the first regime, the radioactive solutions from the chromate holes did not occur in the Techa River.

Releases from the chromate holes into the Techa River after preliminary settling (regime No. 2) were performed in periods from September 1950 to February 1951 (Gromov and Gladyshev 1951) and in September-October 1951 (Alexandrov et al. 1951, Demyanovich et al. 1952b). According to technological process, the time for settling of product 922 was to be equal to the time required for complete precipitation of barium chromate and barium carbonate with the capture of radioactive strontium (Ratner et al. 1950). At this, the solution released into the Techa River would only contain Cs and Ru and less than 0.7% of Sr (Ratner et al. 1950). However, the time required for complete precipitation of barium chromate and barium carbonate was not always maintained. This fact was already discussed in December 1950 (Minutes, 1950). Therefore, the radionuclide composition of the releases into the Techa River from the chromate hole at the times, corresponding to the regime of settling, differed from the radionuclide composition of non-separated mixture of FP and depended on the quality of alkaline precipitation and the time of settling of product 922 in the chromate hole.

For additional settling of LRW released into the Techa River before their reaching the closest village to the site of the release (Metlino village located 5–7 km downstream), specialists of IPCAS recommended “reconstruction of a pond on the Techa River at the distance of 3 km from the site of releases that will extend the time for settling of the activity adsorbed on dredge” (Ratner et al. 1950). However, this recommendation was only followed in August 1951 when Koksharovsky Pond with the capacity of 400,000 m³ was constructed (Alexandrov et al. 1951). The time for water settling in Koksharovsky Pond was equal to 2 days (Ilyin 1952). With this, the specific activity of water decreased due to radioactive decay, sedimentation of suspended particles and sorption of the radionuclides by bottom sediments.

In conclusion of the analysis of the technological processes for LRW management at MPA during the first period of its operation it can be stated that:

- Increase in plutonium production in 1949-1951 continuously accompanied with the increase in the amount of LRW released from Facility B into the Techa River. Technology for LRW reprocessing changed for several times that led to changes in the activity and radionuclide composition of the releases.
- In 1951, accidental situations occurred several times in the departments of Facility B responsible for storage and reprocessing of LRW (overflow of the chromate hole in Department 16, leaks of HLW with cooling waters in Complexes C). At this times, the amount of the activity released into the Techa River sharply increased.

3. ANALYSIS OF DATA ON SPECIFIC ACTIVITY AND RADIOCHEMICAL COMPOSITION OF LRW

This section summarizes data on assessment of specific activity and radiochemical composition of the main types of LRW from Facility B provided in archival documents (Perminov et al. 1950; Alexandrov et al. 1951; Demyanovich et al. 1952b; Starik et al. 1952; Ilyin 1956). In order to analyze available data it is necessary to evaluate possibilities of methods for radiochemical analysis and radiation detection used at Mayak PA at that time.

3.1. Methods for radiation detection and radiochemical analysis in 1950-1956

This section provides a short description of methods for radiometry and radiochemical analysis used at the Central Plant Laboratory (CPL) of Mayak PA in the beginning of 1950s as originally described in Doctoral Thesis of D.I. Ilyin (1956). Measurements were also performed at Analytical Laboratory of Facility B. Unfortunately, detailed description of methods used at the Analytical Laboratory could not be found. It could only be supposed that these methods were analogous to those used at the CPL. It should also be noted that the section includes methods for measurement of beta- and gamma-radiation since we only consider uranium fission products (methods for measurement of alpha-radiation are not considered).

3.1.1. Methods for measurement of gamma-radiation

Gamma-radiation of a sample was measured with Geiger-Muller cylindrical counter via comparison with gamma-radiation of a standard source (^{60}Co) by a number of counts per minute at the same geometric conditions of measurement. Measurements were conducted with counter AMM-4; the counter walls were made of glass of 1-mm thickness covered with a layer of copper. The counter was placed into a lead housing that had a window for photons penetration along the counter length. The counter was surrounded with aluminum and lead filters with the thickness of 1.6 mm and 5 mm, respectively. The lead filter absorbed beta-particles and low-energy photons from a sample. The aluminum filter absorbed photoelectrons, Compton-electrons and electron-positron pairs, broken up from lead by photons. The cobalt standard sources were calibrated with radium standard (in mg-equ Ra) each 15 days. There were 5 pairs of liquid cobalt standard sources in the range from 0.001 to 5 mg-equ Ra. Radiometry of sample and the standard source (both were as liquids in test-tubes; the volume of solution was 5 cm³) was conducted at five fixed distances from the counter; the distance ranged from 7 to 120 cm. The error of the method according to the estimates of authors was 10%.

It is known that sensitivity of gas counters depends on the energy of detected radiation, especially in a low-energy range up to 0.6 MeV (Ivanov 1970). Energy sensitivity of a counter is expressed by a ratio of a counting rate to exposure dose-rate. The ratio, in turn, is proportional to a ratio of the energy, absorbed in detector, to emitted energy.

To evaluate how the energy sensitivity could influence the results of measurement of gamma-activity of different radionuclides from the mixture of fission products, we applied numerical Monte Carlo modeling to describe energy absorption in detector AMM-4. The geometry of measurements was described in detail and can be reproduced in numerical modeling performed with the use of MCNP4C-b code. The source was modeled as an isotropic emitter of mono-energetic photons, from the energy of 0.1 MeV, uniformly distributed in a test tube with water (imitation of a typical sample). Analogous modeling allowed calculation of the energy absorbed in the detector from original photons and secondary radiations.

The counter was calibrated using ^{60}Co as a standard, thus, taking the ratio of calculated absorbed energy in the detector to the mean emitting energy of ^{60}Co photons (1.25 MeV) equal to unity, the energy sensitivity of the counter can be expressed through relative ^{60}Co -equivalent units (Fig. 4).

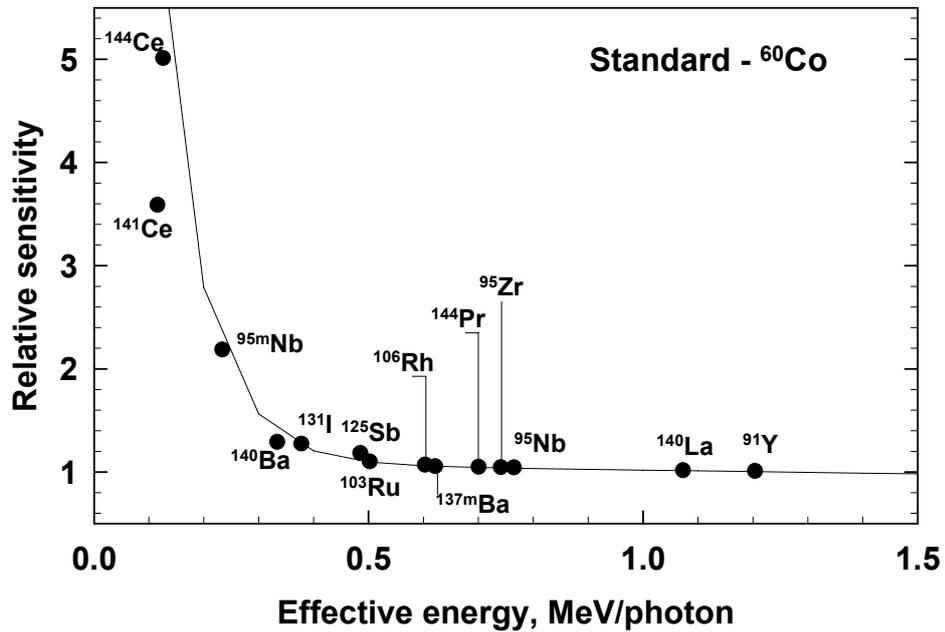


Fig. 4. Energy sensitivity of Geiger-Muller gas counter with a copper cathode (AMM-4). The line represents theoretical dependence, obtained by numerical modeling of mono-energetic photons. Points indicate sensitivity to radiation of different radionuclides with specific mean effective energies.

It is seen from Fig. 4, that for gamma-emitters with low photon energy (such as ^{141}Ce and ^{144}Ce), measurements made with the counter calibrated using high-energetic gamma-radiation of ^{60}Co , could be 3-5 times higher than the true values. On the other hand, the lead filter completely absorbed low-energy gamma-radiation of ^{141}Ce and ^{144}Ce . As a result, measurement of samples with determinant amounts of ^{141}Ce and ^{144}Ce , as well as other radionuclides with low-energy gamma-radiation, could have significant systematic error. At present, our studies of detection methods with the use of numerical modeling continue.

3.1.2. Methods for measurement of beta-radiation

Beta-particles in a sample were detected with the use of so called "Device B" with an end-window counter that had a thin mica window with the thickness not greater than 5 mg/cm^2 (so beta-particles could enter the counter). The device was calibrated with the use of standard sources made of uranium oxide; calibration was performed monthly and each time when the counter was replaced. Calibration was performed using a filter with the thickness of 0.046 g/cm^2 to cut off contribution of beta-radiation of the standard source $\leq 0.2 \text{ MeV}$. The counting coefficient was determined from measurements of five standard sources with the accuracy of 1% for each source. At the same time, correction for background and gamma-radiation of a standard source was made. The counting coefficients were determined at 3-5 fixed positions of a standard source. Preparation of a sample depended on its specific activity and salt composition: a sample was either evaporated by 10-100 times, or diluted. For measurement, solutions of a volume of $0.25\text{-}1.0 \text{ cm}^3$ were applied on two substrates of aluminum foil with a hollow with diameter of 38 mm. Both samples were first

measured without filters. Then, one of them, was measured with four aluminum filters with the thickness from 2 to 16 mg/cm². Besides this, gamma-radiation background of a sample was measured, for this, a filter with a thickness of 2 g/cm² was placed between a sample and the counter, so the filter completely absorbed all beta-particles. Measurements of a sample with different filters were used to assess correction for the thickness of mica window and air layer between a sample and the counter. The correction was accounted for at determination of beta-radiation of an original solution that was assessed from measurements of both samples without filters. Besides this, if a sample was quite thick, correction for self-absorption was also made. Accuracy of this method was 3-5%. Sometimes, a simpler “relative” method was used that did not measure corrections individually for each sample. Accuracy of the relative method did not exceed 15% according to estimates of the authors.

The question on possible systematic errors of beta radiometry was considered in Mokrov (2003). According to the author, radiometry of activity via detection of counting rate of beta-particles can be underestimated for ⁹⁵Nb, ¹⁰³Ru and ⁹⁵Zr due to absorption of low-energy beta-radiation in a mica window of an end-window counter and in air between a measured sample and window of the counter. It should be noted that the end-window counter, described by Ilyin (1956), had sufficiently thin window, so the systematic error could not have significant contribution in results of measurements. However, an end-window used in 1950 and described in Perminov et al. (1950) could have significant systematic error at measurement of ⁹⁵Nb, ¹⁰³Ru and ⁹⁵Zr.

At present time, we continue analysis of possible systematic errors in measurement of beta-radiation with the use of Monte Carlo numerical modeling of energy absorption in detectors used at Mayak PA in 1950-1956.

3.1.3. Radiochemical methods for determination of specific radionuclides

Chemical methods available in the beginning of 1950s allowed extraction of five groups of radionuclides from original mixture of fission products. They are:

1. Radioactive cesium (¹³⁷Cs);
2. Alkaline earth elements, AEE (⁹⁰Sr, ⁸⁹Sr, ¹⁴⁰Ba);
3. Rare earth elements, REE (¹⁴¹Ce, ¹⁴⁴Ce, ⁹¹Y);
4. Radioactive ruthenium (¹⁰³Ru, ¹⁰⁶Ru);
5. Sum of zirconium and niobium (⁹⁵Zr, ⁹⁵Nb).

Here we do not provide detailed description of chemical methods for extraction presented in Ilyin (1956). Contribution of particular groups of radionuclides in total activity was evaluated as the ratio of beta- or gamma-radiation of extracted fraction to beta- or gamma-radiation of initial solution, respectively. It is evident that such evaluation significantly depended on possible instrumental errors in radiometry as well as effectiveness of chemical methods for extraction of particular groups of radionuclides from initial solution. Beside this, partial contribution of fractions of gamma- and beta-emitters could also be different (as an example, there are two radiochemical compositions of products 757 and 901 in the report of Demyanovich et al. 1952b: One composition was obtained from gamma-emitters, the other – from beta-emitters). In most analyzed documents, radiochemical composition of LRW was evaluated from beta-emitters, due to the fact, that the AEE group contained only pure beta-emitters after a fast decay of ¹⁴⁰Ba. Method for strontium determination does not contain description how the chemical outcome was determined (amount of co-precipitation on a carrier). Description is not provided whether particular fractions were stored for the time necessary for establishment of equilibrium between parental and daughter radionuclides

after chemical extraction before radiometry measurements. The notes above provide evidence that radiochemical methods used in the beginning of 1950s could have significant errors that should be taken into account in data analysis.

3.2. Assessment of specific activity and radiochemical composition of technological releases

This section includes characteristics of technological LRW that were released into the Techa River and played significant role in contamination of the river water and bottom sediments at different time periods.

3.2.1. Alkaline decantate from Department 7 (product 757)

Alkaline decantate from department 7 (product 757) was classified as low-level activity and released into the Techa River since the beginning of facility B operation. From the end of December 1948 to December 1949 it was discharged into the Techa River after adsorptive purification (Ratner et al. 1950) that resulted in decrease of initial activity by 8-10 times. From January 1950 to October 28, 1951, product 757 was discharged into the Techa River without purification (Ratner et al. 1950; Alexandrov et al. 1951). From October 28, 1951, release of product 757 was switched to Karachay Lake. Available estimates of specific activity of product 757 are shown in Table 2.

Table 2. Specific activity of product 757 according to different archival sources

Source	Specific activity		Comments
	mg-equ Ra/L	mCi/L	
Ratner et al. 1950	5 – 10	-	Method of evaluation is not indicated
Alexandrov et al. 1951	1.4	-	Method of evaluation is not indicated
Alexandrov et al. 1951	0.8	-	Measurements of September 25-October 5, 1951
Demyanovich et al. 1952b	0.45 (0.37 – 0.61)	1.5 (1.2 – 1.9)	Measurements of March 3-31, 1952
Starik et al. 1952	0.5	1.0	Based on measurements

It is seen from Table 2, that estimates of specific activity derived from real measurements are in the range from 0.5 to 0.8 mg-equ Ra/L and from 1.0 to 1.5 mCi/L. Daily average debit of product 757 was estimated to be equal to 70 m³/d in September-October 1951 (Alexandrov et al. 1951) and 90 m³/d in April 1952 (Starik et al. 1952).

Estimates of radiochemical composition of product 757 are given in Table 3. It is seen from Table 3, that according to CPL data, the main contribution into specific activity of product 757 in 1951 was from radioactive strontium (AEE group); cesium was absent in the solution. However, measurements performed in 1952 showed that from 50 to 80% of total activity of product 757 was due to ruthenium group; the contribution of strontium was about 6% and the contribution of cesium varied from 6% to 11%. Such difference in the estimates is possibly related to the fact that the technology for Pu extraction at facility B was substantially changed in 1952 (fluoride and extraction technology was substituted by a technology for extraction and purification of Pu). For this reason,

estimate of 1951 (Alexandrov et al. 1951), seemingly, better describes the radionuclide composition of product 757 at the time of its release into the Techa River.

Table 3. Radiochemical composition of product 757 according to different archival sources

Source	Contribution of the group of radionuclides, %					Comments
	AEE	Cs	Ru→Rh	REE	Zr→Nb	
Alexandrov et al. 1951	59	0	35	2	4	Reference to CPL data
Demyanovich et al. 1952b	6.1	11.1	51.3	0	31.5	Measurements of March 3-31, 1952
Starik et al. 1952	5.7	5.7	79.6	0.6	7.4	Evaluated in 1952 from the results of measurements

3.2.2. Decantate from Department 3 after chromate purification (product 922)

Product 922 appeared since May 1950, when the reprocessing of decantates of Department 3 (product 901) commenced with the use of barium and carbonate precipitation in the chromate hole (Ratner et al. 1950). During the first 4-5 months of operation of the chromate hole, product 922 did not enter the Techa River from the hole. From September-October 1950 to the beginning of operation of the second chromate hole (approximately in April-May 1951) product 922 was released into the Techa River after settling in the chromate hole that almost stopped filtrating. It is supposed, that during the first several months the second chromate hole worked in filtration regime and product 922 did not enter the Techa River (as in the case with the first chromate hole). However, on September 17, 1951, the second chromate hole stopped working and product 922 was released into the Techa River after settling in the first chromate hole.

Available estimates of specific activity of product 922 are provided in Table 4.

Table 4. Specific activity of product 922 according to different archival sources

Source	Specific activity		Comment
	mg-equ Ra/L	mCi/L	
Perminov et al. 1950	10 (7 – 12)	–	Method of evaluation is not indicated. It is noted, that about half of activity is in precipitate; thus, taking this into account, specific activity of solution is equal to 3-3.5 mg-equ Ra/L
Alexandrov et al. 1951	12	20	Method of evaluation is not indicated
Demyanovich et al. 1952b	11 (8.5 – 28)	30 (16 – 70)	Measurements of October 9-26, 1951 after passage of product through adsorbers
Starik et al. 1952	7	18.5	Evaluated from measurements

It is seen from Table 4, that different archival sources give similar estimates of average specific activity of product 922: from 7 to 12 mg-equ Ra/L and from 20 to 30 mCi/L. These levels are significantly lower than the specific activity of initial product 901 that was equal to 20-35 mg-equ Ra/L and 100-120 mCi/L. Daily average debit of product 922 was evaluated to be 35-45 m³/d

in May-July 1950 (Perminov et al. 1950), 100 m³/d in September-October 1951 (Alexandrov et al. 1951) and 175 m³/d in April 1952 (Starik et al. 1952). Increase in the daily debit reflects increase in plutonium production at Facility B. Estimates of radiochemical composition of product 922 are provided in Table 5.

Table 5. Radiochemical composition of product 922 according to different archival sources

Source	Contribution of the group of radionuclides, %					Comments
	AEE	Cs	Ru→Rh	REE	Zr→Nb	
Perminov et al. 1950	1	40	59	-	-	Design data
Perminov et al. 1950	1-4	56-64	25-26	-	-	Laboratory experiments
Alexandrov et al. 1951	1.8	50.5	47.7	0	0	Design data
Starik et al. 1952	23.2	21.6	49.4	3.6	2.2	Evaluated in 1952 from measurements

It is seen from Table 5, that according to the design, product 922 was to be completely purified from radioactive strontium and its activity to be only determined by radionuclides of cesium and ruthenium. However, measurements of real solutions in 1952 showed that about 20% of specific activity of product 922 was contributed by AEE group. It should be noted that since 1952, barium-carbonate purification of product 901 was conducted with the use of apparatus of a new Department 19 at Facility B (this Department did not exist in 1950-1951); after purification it was released in a closed technological reservoir – Karachay Lake. Therefore, data of 1952 do not reflect changes in radiochemical composition of this type of LRW during its settling in the chromate hole. We note, that radiochemical composition of product 922 evaluated in 1952 from the results of measurements is quite similar to the composition of original product 901 (Table 6). Therefore, reprocessing of product 901 in 1952 only resulted in decrease in specific activity from 100-120 mCi/L to 20-30 mCi/L (Demyanovich et al. 1952b).

Table 6. Radiochemical composition of product 901 according to measurements performed on March 3-31, 1952 (Demyanovich et al. 1952b).

Contribution of the group of radionuclides, %				
AEE	Cs	Ru→Rh	REE	Zr→Nb
20.9	28.9	41.6	5.1	3.4
(12 – 28)	(19 – 35)	(13 – 50)	(1 – 6)	(0.1 – 5.6)

In 1950-1951, radiochemical composition of product 922 depended on the time of settling in the chromate hole. However, archival documents noted in December 1950, that settling of product 922 from precipitate of barium chromate and barium carbonate was not sufficient because the settling time had to be reduced due to large amounts of reprocessed products (Minutes, 1950). For these reasons, it can be concluded that radiochemical composition of product 922 released into the Techa River varied significantly dependent on the quality of barium-carbonate purification and settling time in the chromate hole.

3.2.3. Solution of manganous pulp in bisulphite (product 91/92)

The start of release of product 91/92 corresponded to the launch of Department 15 where uranium was obtained (approximately in the end of 1950). Available estimates of specific activity of product 91/92 are given in Table 7.

Table 7. Specific activity of product 91/92 according to different archival sources

Source	Specific activity		Comment
	mg-equ Ra/L	mCi/L	
Alexandrov et al. 1951	-	143	Method of evaluation is not indicated
Starik et al. 1952	225	112	Evaluated from measurements

It is seen from Table 7, that different archival documents provide similar estimates of specific activity of product 91/92. Daily average debit of product 91/92 was evaluated to be 2.8 m³/d in September-October 1951 (Alexandrov et al. 1951) and 1.0 m³/d in April 1952 (Starik et al. 1952). Decrease in daily debit is explained by the fact that at the initial period after the launch, Department 15 had to reprocess all solutions accumulated in storages of Department 15a since the beginning of facility B operation. Starik et al. (1952) noted that remained solutions were reprocessed in 1952. Estimates of radiochemical composition of product 91/92 are provided in Table 8.

Table 8. Radiochemical composition of product 91/92 according to different archival sources

Source	Contribution of the group of radionuclides, %					Comment
	AEE	Cs	Ru→Rh	REE	Zr→Nb	
Alexandrov et al. 1951	0	0	0	0	100	Design data
Starik et al. 1952	0	0	0.5	0	97.5	Evaluated in 1952 from measurements

It is seen from Table 8, that according to the design, activity of products 91/92 was completely determined by radionuclides of zirconium and niobium. Measurements of real solutions in 1952 confirmed the design data.

3.2.4. Aluminate waters (product 673)

Release of product 673 into the Techa River commenced since December 20, 1950 (Minutes 1950). The product was released without purification. Before the date, at different time periods, the product was either conveyed to Staroe Boloto or reprocessed in chromate hole together with product 922 (Perminov et al. 1950; Minutes 1950). Available estimates of specific activity of product 673 are given in Table 9. Analysis of data from Table 9 suggests, that estimate of specific activity from Perminov et al. 1950 is slightly overestimated since the other three archival documents give quite similar estimates: 1-3 mg-equ Ra/L and 3-10 mCi/L. Daily debit of product 673 was evaluated to be equal to 50 m³/d in December 1950 (Minutes 1950) and 16-23 m³/d in 1952 (Starik et al. 1952; Ilyin 1956). The reason for the decrease in daily debit is not clear, it seems, that it could be due to changes in technology for reprocessing of this type of LRW at facility B in 1952.

Table 9. Specific activity of product 673 according to different archival sources

Source	Specific activity		Comment
	mg-equ Ra/L	mCi/L	
Perminov et al. 1950	15	-	Method of evaluation is not indicated
Minutes 1950	1.0	-	Method of evaluation is not indicated
Starik et al. 1952	1.0	2.5	Evaluated from measurements
Ilyin 1956	3.0	10	Reference to reports of Laboratory of Facility B of 1952 and 1955

Estimates of radiochemical composition of product 673 are presented in Table 10. It is seen from Table 10, that estimates of radiochemical composition of product 673 are close.

Table 10. Radiochemical composition of product 673 according to different archival sources

Source	Contribution of the group of radionuclides, %					Comments
	AEE	Cs	Ru→Rh	REe	Zr→Nb	
Alexandrov et al. 1951	9.3	-	44	26.7	20	Design data
Starik et al. 1952	7	7	48	19	20	Evaluated in 1952 from measurements

3.3. Assessment of specific activity and radiochemical composition of non-technological releases

This section includes characteristics of LRW that were not foreseen in initial design of facility B, however, the release of these types of LRW into the Techa River was regular and could play significant role in contamination of the river water and bottom sediments.

3.3.1. Condensate from the diffuser of the main stack

This type of LRW was not accounted for in the initial design because it was supposed that the draught in the stack will be sufficient enough to release all relief gases into the atmosphere (Gladyshev 1992, p. 28-29). However, it was revealed in Summer 1950, that a condensate of vent gases settled on internal walls of the concrete pipe and flowed down into workrooms of Facility B. (Gladyshev 1992, p. 28-29). For this reason, in August 1950, a stainless steel pipe was constructed inside the concrete pipe for relief gases from technological apparatus. The condensate was diverted into special pools and since September 1950, the CDMS was conveyed to Department 16 and subsequently discharged into the Techa River. Data on any purification of the CDMS in Department 16 are not available in archive documents that we analyzed. It could be supposed from indirect data that its release into the Techa River was ceased in November 1951 (it is followed from Demyanovich et al. (1952b), that in January 1952 the CDMS was released into the chromate hole). Available estimates of specific activity of the CDMS are given in Table 11.

Data from Table 11 show high variability in specific activity of the CDMS amounting to two orders of magnitude. First of all, it should be noted, that the activity of aerosols in the pipe

significantly changed during a day: Solution of blocks in Department 2 resulted in an increase of aerosols by 10-20 times (Alexandrov et al. 1951). For this reason, frequency and methods of sampling played significant role in evaluation of specific activity of the condensate. Also, it can be assumed, that formation of the condensate was higher in winter months at low external temperatures than in warm periods of a year. For this reason, measurements from Demyanovich et al. (1952b), conducted in the middle of winter, could be considered as maximum estimates. It should be noted, that at this period, gamma-activity was determined from 17 samples but beta-activity – only from 8 samples and the median value of beta-activity is 6 times lower than the average value. Therefore, the data are characterized by a high uncertainty.

Table 11. Specific activity of the condensate from the diffuser of the main stack according to different archival sources

Source		Specific activity		Comments
		mg-equ Ra/L	mCi/L	
Demyanovich et al. 1952b	Average	62	262	Measurements of January 1 – February 10, 1950. The CDMS released in the chromate hole
	Median	14	43	
	Range	(6 – 456)	(32 – 850)	
Starik et al. 1952		40	71	Evaluated from measurements ¹
Ilyin 1956		2.8	8	Reference to reports of Laboratory of Facility B of 1952 and 1955

Starik et al. (1952) give estimate for a mixed solution that beside CDMS contained condensates of ejectors of Department 2 and multicyclones of Department 3 as well as drainage and washing solutions from communication line A-206

Estimates of daily debit of the CDMS are only available for the period of the beginning of 1952 and different documents provide quite similar values: 25-30 m³/d (Demyanovich et al. 1952b), 20 m³/d (Starik et al. 1952) and 25 m³/d (Ilyin 1956). Estimates of radiochemical composition of the CDMS are given in Table 12.

Table 12. Radiochemical composition of the CDMS according to different archival sources

Source	Contribution of the group of radionuclides, %					Comments
	AEE	Cs	Ru→Rh	REE	Zr→Nb	
Starik et al. 1952	16.7	0.5	6	43.4	33.4	Evaluated from measurements ¹
Ilyin 1956	9.1	1.0	4.8	60	24.8	Reference to reports of Laboratory of Facility B of 1952 and 1955

Starik et al. (1952) give estimate for a mixed solution that beside CDMS contained condensates of ejectors of Department 2 and multicyclones of Department 3 as well as drainage and washing solutions from communication line A-206

It is seen from Table 12, that different archival sources give similar estimates of the radiochemical composition of the CDMS. It is evident, that composition of the mixture in this product should be close to theoretical composition of FP with the age equal to hold-up time. Ratio

of contribution of AEE group to cesium-137 in estimates of Starik et al. (1952) for the beginning of 1952 corresponds to that of FP of the age of 70-80 days and is equal to hold-up time in this calendar year (see Fig. 1).

3.3.2. Desorption waters from apparatus and canyons

Desorption purification of apparatus was not planned in the initial design because it was not expected that significant amount of radioactive “deposits” would be formed on the walls of apparatus and service lines. These waters were extremely contaminated with chemical substances, because to dissolve “deposits” on the walls of the apparatus, solutions of potassium permanganate, caustic soda, nitric, oxalic, and even hydrofluoric acids were used (Ilyin 1956). This type of LRW contained notable amounts of U and Pu and were to be conveyed to tanks C. However, according to Alexandrov et al. (1951), “there were cases of their release into the Techa River without preliminary analysis”. According to Starik et al. (1952), release of this type of LRW into the Techa River continued in April 1952. Available estimates of specific activity of desorption waters are given in Table 13.

Table 13. Specific activity of desorption waters according to different archival documents

Source	Specific activity		Comments
	mg-equ Ra/L	mCi/L	
Demyanovich et al. 1952b	11	58	Estimated from measurements of February 17-April 30, 1952 Solutions from line A-317
	(0.65 – 54)	(1.5 – 274)	
Starik et al. 1952	0.37	1.4	Solutions from line A-206
	(0 – 1.2)	(0.03 – 3.8)	
Ilyin 1956	23.6	50	Estimated from measurements of solutions from line A-317
	5.4	13	
			Reference to reports of Laboratory of Facility B of 1952 and 1955

Demyanovich et al. (1952b) provide the results of measurements of solutions from two service lines: line A-317 collected “waters from washing of canyons, surfaces of apparatus and, partially, products from desorption of apparatus”, and line A-206 collected “solutions from desorption of apparatus and their parts”.

Data from Table 13 show a very large variability in estimates of specific activity of desorption waters. It should be noted, that according to Ilyin (1956), activity of desorption waters significantly decreased with washing of an apparatus (from 10^{-2} Ci/L to 10^{-7} Ci/L). It was noted in Demyanovich et al. (1952b), that waters from desorption of apparatus had, as a rule, significant amounts of non-soluble precipitates, mainly containing MnO_4 , SiO_2 and $PbSO_4$, with an activity that was 10 times higher than a solution. Since sampling of an average product sample, containing significant amounts of non-soluble precipitates, was a very difficult task, authors of the report (Demyanovich et al. 1952b) recommended considering the results of their measurements as “sufficiently approximate”. They also noted, that radiochemical analysis of this type of LRW (Table 14) was performed, as a rule, only in liquid form.

Table 14. Radiochemical composition of desorption waters according to different archival sources

Source	Contribution of the group of radionuclides, %					Comments
	AEE	Cs	Ru→Rh	REE	Zr→Nb	
Starik et al. 1952	22.1	0.7	7.4	51.5	18.4	Solutions A-317
Demyanovich et al. 1952b	-	-	3.6-4.6	6.9-7.6	88-90	Solutions A-317
	-	-	2-3	-	95-97	Solutions A-206
Ilyin 1956	7.3	0.9	2.7	40	49.1	Reference to reports of Laboratory of Facility B of 1952 and 1955

It is seen from Table 14, that different archival documents provide different estimates of radiochemical composition of desorption waters. According to Demyanovich et al. (1952b), specific activity of this type of LRW was mostly determined by ^{95}Zr and ^{95}Nb ; however, at the same time, the results from Starik et al. (1952) show that more than a half of the activity was due to REE. Data on daily debit of this type of LRW are also discrepant. According to Demyanovich et al. (1952b) and Starik et al. (1952), average debit in the beginning of 1952 was evaluated to be $5 \text{ m}^3/\text{d}$ (ranged from 1 to $15 \text{ m}^3/\text{d}$). Ilyin (1956) gave significantly larger value of $50 \text{ m}^3/\text{d}$.

Therefore, there is a large uncertainty in specific activity and radiochemical composition of desorption waters. Moreover, the frequency of washing of apparatus and canyons is unknown to us. The regularity of the release of this type of LRW is also unknown. It should be noted that if desorption waters were purified through the adsorbers before their release into the Techa River, then the contribution of this type of LRW into total contamination, seemingly, was not large.

3.4. Assessment of radiochemical composition of total LRW release from Facility B

Estimates of radiochemical composition of total release of LRW from Facility B in the beginning of 1950s are available from three archival documents (Alexandrov et al. 1951; Starik et al. 1952; Ilyin 1956). These estimates are given in Table 15.

Table 15. Radiochemical composition of the total release from Facility B according to different archival sources

Source	Contribution of the group of radionuclides, %				
	AEE	Cs	Ru→Rh	REE	Zr→Nb
Alexandrov et al. 1951	19.9	2.7	7.8	48	21
Starik et al. 1952	20.8	12.6	32.5	19.3	16.6
Ilyin 1956	20.4	12.2	25.9	26.8	13.6

It is seen from Table 15, that estimates of radiochemical composition, obtained by Starik et al. (1952) and Ilyin (1956) are in general agreement with each other, but significantly different from estimates of Alexandrov et al. (1951) on contribution of Cs, Ru and REE. Regarding the contribution of strontium group into total activity, all three archival documents give similar values close to 20%; estimates on the contribution of $^{95}\text{Zr}+^{95}\text{Nb}$ (14-21%) can also be considered as comparable.

Analysis of data, shown in Table 15, should be accounted for the following facts. Estimates of Alexandrov et al. (1951) were obtained from the results of radiochemical analysis of the only one sample (taken on September 24–25, 1951). The authors (Alexandrov et al. 1951) wrote:

“This radionuclide composition [Ed. Note: shown in Table 15] is rough because it could be different on different days, however it can be used only for approximate estimates.”

Radiochemical composition from Alexandrov et al. (1951) and Starik et al. (1952) describes only regular releases from Facility B and does not include accidental releases with HLW from Complexes C. Alexandrov et al. (1951) noted that occurrence of products from tanks C in the releases resulted in an increase in contribution of strontium. From Ilyin (1956) it cannot be unambiguously concluded whether his estimates reflected the composition of total release or only described regular release. The first conclusion can be supported by the fact, that the author obtained his estimates not only from measurements of LRW but also from monitoring of radionuclide contamination of water and bottom sediments in Koksharovsky and Metlinsky Ponds and in the Techa River up to its entry into Iset' River (Ilyin 1956).

Besides this, estimates from (Alexandrov et al. (1951) and Ilyin (1956) concern period of massive releases of LRW into the Techa River (1950-1951) and estimates of Starik et al. (1952) are related to the later period (1952), when the technology for obtaining of Pu and LRW management at Facility B changed compared with the previous period.

For these reasons, it is absolutely impossible to make unambiguous conclusion in favor of any estimate of the radiochemical composition of the total release. However, estimates of Starik et al. (1952) and Ilyin (1956), obtained from analysis of many data can be considered as expert estimates of the most typical radiochemical composition of the total release, while Alexandrov et al. (1951) give the results of the single analysis and do not pretend to anything more.

4. RECONSTRUCTION OF THE DYNAMICS OF RELEASES FROM FACILITY B INTO THE TECHA RIVER

During the first years of MPA operation, the releases from Facility B were delivered into three water systems: Techa River, Staroe Boloto and Karachay Lake (since October 28, 1951). Radioactive releases during the period of interest increased in accordance with the increase in capacity of Facility B. The control for radioactive releases from Facility B commenced in August 1951 (Alexandrov et al. 1951). Initially, only gamma-activity of releases from different effluent canals was measured that did not provide evaluation of the levels of contamination with beta-radionuclides (^{89}Sr and ^{90}Sr). Systematic control for the discharges released through the effluent canal (from Facility B into the Techa River) commenced only on September 25, 1951. Since this date, the results of the total activity released from Facility B are available (Alexandrov et al. 1951; Demyanovich et al. 1952b). Measurements of specific activity of the Techa River water directly at the site of releases, at the point of complete mixture of the releases with the river water (so called “point 18”) and downstream in the clearing ponds (Koksharovsky and Metlinsky Ponds) commenced in July 1951. Since specific activity of river water was in close correlation with the amount of release, measurements of specific activity of the Techa River water performed in the upper Tcha-region can be regarded as a part of the monitoring of radioactive releases.

4.1. The results of monitoring performed in 1951-1952 and evaluation of the releases for the period from July 1951 to September 1952

In September 1951, a flow-meter and a sampling device were installed in the pipe, that exits the pumping station and joins the pipe (with a diameter of 500 mm) going directly into the Techa

River, in order to control releases into the river. Each hour, a 250-mL sample of the releases was taken from the pipe and put into a bottle. Thus, 24 samples were taken during a day with a total volume of 6 L. Once a day, content of the bottle was thoroughly mixed and a daily-averaged sample was taken from the bottle for measurements. Total monthly release into the Techa River evaluated from these measurements for the period since October 1951 (Table 31 from Demyanovich et al. 1952b) is shown in Fig. 5. The figure also includes monthly-averaged values of specific activity of river water at the point of complete mixture since July 1951 (Table 21 from Ilyin 1956). High correlation coefficient between these values ($R=0.997$) allowed reconstruction of the monthly release in July-September 1951 with the use of data on specific activity of river water at point 18 (Fig. 5).

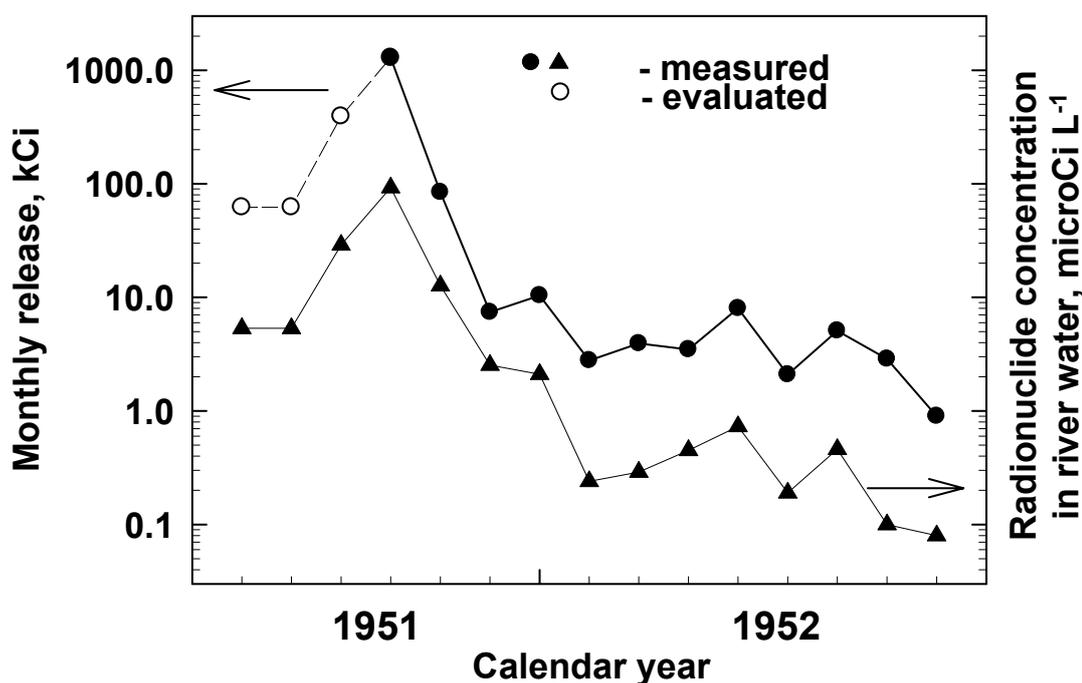


Fig. 5. Total monthly release into the Techa River (Table 31 from Demyanovich et al. 1952b) and monthly-averaged values of specific activity of the river water at the point of complete mixture (Table 21 from Ilyin 1956). Total monthly release in July-September 1951 was reconstructed with the use of corresponding data on specific activity of river water.

It is seen from Fig. 5, that the total release was maximum in October 1951 and amounted to 1.3 million Ci per month (Demyanovich et al. 1952b). This report also provides ten-day data on the activity released into the Techa River. These data provide evidence that the main part of the maximum release occurred during the first and the second 10-day periods of October 1951 (601 and 625 kCi, correspondingly). Since these numbers significantly exceed all other estimates (Alexandrov et al. 1951; Ilyin 1956), we evaluated the balance between “routine” and “accidental” releases for the end of September and the first 10-day period of October 1951. For making such evaluation we used daily data on the activity released into the Techa River from September 25 to October 7, 1951 (Table 1 from Alexandrov et al. 1951). According to these data, represented in Table 16, “the amount of radioactive substances released into the Techa River varied sharply from 82,600 to 710 Ci day⁻¹” during the period of Commission work (Alexandrov et al. 1951). Alexandrov et al. (1951) indicate that “evaluation of the average release into the Techa River was

performed omitting the “wild overflow” of 27–29 September 1951 since it was supposed that such high radioactive releases occurred relatively rare and were not directly connected with the technological processes”. Averaging of the measurements presented in Table 16, except the ones performed on 27–29 September, results in a daily average release for the 10-day period equal to 4,100 Ci day⁻¹.

Table 16. Total release of radioactive wastes into the Techa River in September–October 1951 (data are from Alexandrov et al. 1951).

Date	Volume, m ³	Specific activity		Daily release	
		mg-equ Ra L ⁻¹	mCi L ⁻¹	g-equ Ra	Ci
25 Sept	8,350	0.525	1.09	4,383	9,100
26 Sept	8,350	0.354	0.71	2,950	5,930
27 Sept	8,100	5.5	10.2	44,500	82,600
28 Sept	8,300	-	9.0	-	74,700
29 Sept	8,125	-	3.4	-	27,500
30 Sept	7,850	0.21	0.6	1,650	4,710
1 Oct	7,850	0.051	0.09	392	710
2 Oct	8,070	0.82	1.0	6,620	8,070
3 Oct	8,090	0.052	0.1	421	809
4 Oct	8,900	0.052	0.22	463	979
5 Oct	8,600	0.17	0.53	1,462	4,560
6 Oct	9,200	0.105	0.35	966	3,220
7 Oct	8,800			924	3,080

According to data, outlined in Table 16, during the period of 1-7 October, the release into the Techa River was equal to 21.5 kCi. Taking into account, that the total release for the period of 1-10 October was 601 kCi, the release during three days of 8, 9 and 10 October 1951 was equal to 579.5 kCi that corresponded to daily release of 193.1 kCi day⁻¹. If the value of “routine releases” is taken to be 4.1 kCi day⁻¹ then the value of “accidental releases” will be 189 kCi day⁻¹. In condition of continuous supply of cooling water at the rate of 10-12 m³ hour⁻¹ (Krasnopeyev and Karpov, 1984), specific activity of “wild overflow” of October 8–10 will be equal to 660–790 mCi L⁻¹. This value is in good agreement with the measurement of specific activity of water in the canyon of tank 16 in Complex C-4 performed on October 9, 1951 that was equal to 815 mCi L⁻¹ (Table 3 from Alexandrov et al. 1951). Thus, the maximum value of “wild overflows” in September 1951 was evaluated to be 78.5 kCi day⁻¹ and in October this value increased by nearly a factor of 2.5 and amounted 190 kCi day⁻¹.

Since the specific activity of river water at the point of complete mixture (point 18) was in close correlation with the total release, the estimates of monthly total release in July-September 1951 (Fig. 5) were obtained with the use of linear regression:

$$Y(\text{kCi month}^{-1}) = -12.2 + 13.94 X(\mu\text{Ci L}^{-1}). \quad (1)$$

According to these estimates, monthly releases in July-August constituted 62.2 kCi month⁻¹ with the increase in September up to 392 kCi month⁻¹. These estimates are quite reasonable, because it is known that since September 17 the chromate hole did not provide filtration and the release of product 922 (decantate from Department 3 after precipitation of barium chromate and barium carbonate) increased. However, these estimates cannot be considered as reliable for the following reasons. First of all, daily-averaged values for the period of July-September 1951 from

Ilyin (1956) were obtained on the basis of few water samples. Secondly, the flow-rate should be necessarily taken into account when considering correlation between specific activity of water and the total release. The flow-rate in the Techa River depended on the amount of water released from Kyzyl-Tash Lake through dam D-2 (Fig. 6).

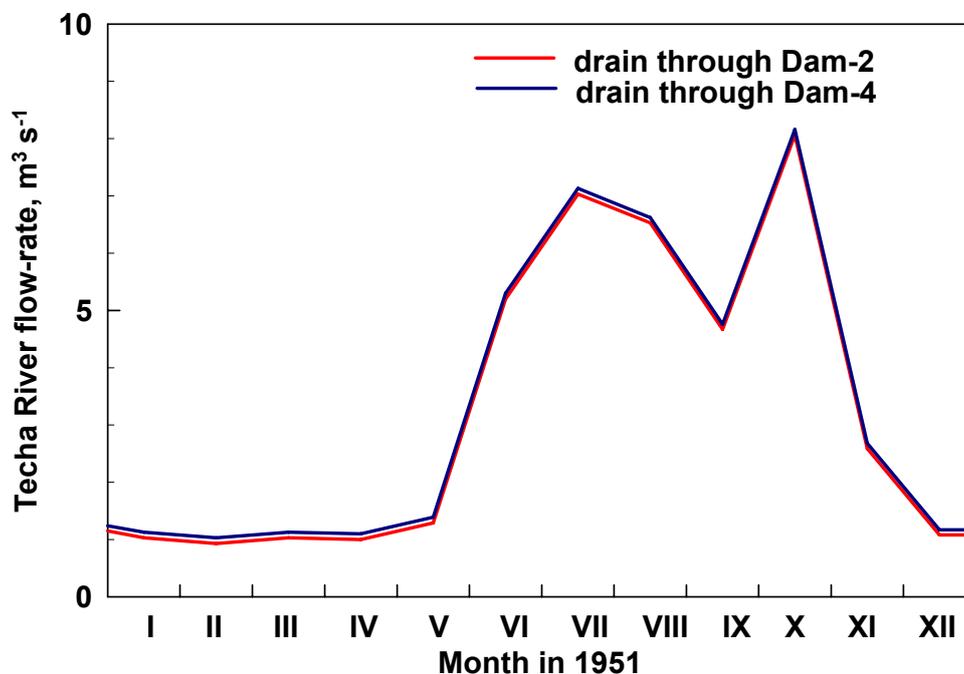


Fig. 6. Monthly flow-rate of the Techa River in 1951 at the point of outflow from Kyzyl-Tash Lake through dam D-2 and at the point of outflow from Metlinsky Pond through dam D-4.

Water release from Kyzyl-Tash Lake was controlled in a wide range (Water balances 1952, Mokrov 2002). According to estimates from Mokrov (2002), flow-rate through dam D-2 was increased in July 1950 from $0.6 \text{ m}^3 \text{ s}^{-1}$ to $1.3 \text{ m}^3 \text{ s}^{-1}$ and in June 1951 up to $5.2 \text{ m}^3 \text{ s}^{-1}$. The period of July-October 1951 characterized by maximum flow-rate through dam D-2: monthly averaged flow-rates were equal to $7.0 \text{ m}^3 \text{ s}^{-1}$ in July, $6.5 \text{ m}^3 \text{ s}^{-1}$ in August, $4.7 \text{ m}^3 \text{ s}^{-1}$ in September and $8.1 \text{ m}^3 \text{ s}^{-1}$ in October (Fig. 6). It was discussed in the previous section, that the increase in flow-rate through dam D-2 was aimed at dilution of LRW released into the Techa River. The time dependence in flow-rate through dam D-4, located at the exit from Metlinsky Pond at the distance of 7 km from dam D-2, corresponded well to that of flow-rate through dam D-2 (Fig. 6).

Since measurements of the specific activity of water in Metlinsky Pond commenced on July 5, 1951 (earlier than measurements in the effluent canal), the authors (Alexandrov et al. 1951; Ilyin 1952) provided estimates of total release through dam D-4 at the exit from Metlinsky Pond. The report of Alexandrov et al. (1951) includes estimates of daily release of activity through dam D-4, obtained from daily measurements of specific activity of water and flow-rate ($7.5 \text{ m}^3/\text{s}$) in July-August 1951 (Table 17). The report of Ilyin (1952) includes estimates of monthly-averaged activity passed through dam D-4 (Table 18).

Table 17. The results of measurements of specific activity of water and estimates of daily release of radionuclides through dam D-4 at the exit from Metlinsky Pond in July-August, 1951 (Alexandrov et al. 1951).

Date	Specific activity of water, $\mu\text{Ci L}^{-1}$	Daily release, Ci day^{-1}
July 5, 1951	63.4	41,080
July 15, 1951	9.0	5,830
July 23, 1951	4.3	2,780
July 24, 1951	3.9	2,500
August 04, 1951	3.9	9,530
August 18, 1951	14.7	7,500

Table 18. Estimates of daily release of radionuclides through dam D-4 at the exit from Metlinsky Pond monthly in 1951–1952 (Ilyin 1952).

Month and year	Daily release, Ci day^{-1}	Month and year	Daily release, Ci day^{-1}
July 1951	7,000	January 1952	141
August 1951	4,100	February 1952	39
September 1951	5,600	March 1952	57
October 1951	NA	April 1952	48
November 1951	1,800	May 1952	31
December 1951	3,015	June 1952	32

Analysis of data presented in Tables 17 and 18 should take into consideration the following facts. The settling time of releases in Metlinsky Pond was 3-5 days, while in Koksharovsky Pond—2 days. Thus, releases from Facility B reached dam D-4 in 3-5 days before August 11, 1951 [when Koksharovsky Pond was created] and in 5-7 days after creation of the Pond. Radioactive decay, sedimentation of suspended particles and sorption of radionuclides with bottoms sediments (BS) occurred in this time frame. On the other hand, by the beginning of June 1951, large amounts of radionuclides had been accumulated in BS in the Techa region from the site of releases to the exit from Metlinsky Pond that could be desorbed back into the river water. According to data from Alexandrov et al. (1951), specific activity of BS measured on August 8, 1951, amounted 20 mCi kg^{-1} and was 200 times higher than maximum levels of the river water contamination. Increased release of clear water through dam D-2 in June-October 1951 led to washout of radionuclides (Fig. 6). Such regime of the clean water release can be considered as a kind of washing of Metlinsky Pond, since the capacity of Metlinsky Pond was 2.5 million m^3 and monthly renew occurred 5-8 times (Mokrov 2002). As a result of the washing, some amounts of BS with high specific activity were captured by water flow and entered the Techa River below dam D-4.

Comparison of the data from Table 18 (radioactive drain through dam D-4) with the results of measurements in the effluent canal of Facility B (Table 31 from Demyanivich et al 1952b) showed that they did not correlate well (the value of the correlation coefficient was low and was equal to $R=0.437$). Thus, measurements of the radioactive contamination of the river water below dam D-4 are not reliable for prediction of the time dependence in the levels of radioactive releases from Facility B. Nevertheless, data from Table 17 show that “wild overflows” exceeding 40 kCi day^{-1} could occur in the beginning of July 1951.

To evaluate the variability in daily release in July-September 1951, we used available data on measurements of specific activity of river water sampled in the Techa-region upstream from dam D-4. The results obtained at the point of complete mixture (point 18) were preferable in our analysis. In case, if data for point 18 were absent we used the data obtained at points located close to point 18 (Table 19). Regression equation (1) accounted for correct units (Ci day^{-1} instead of kCi month^{-1}) was used for evaluation of the release.

Table 19. Specific activity of river water at the point of complete mixture and the estimates of daily release of radionuclides from Facility B in July-September 1951.

Date	Specific activity of river water, $\mu\text{Ci L}^{-1}$	Daily release, Ci day^{-1}
July 5, 1951	100	46000
July 15, 1951	10.2	4300
July 23, 1951	16.8	7400
July 24, 1951	4.0	1450
August 03, 1951	8.0	3300
August 04, 1951	1.75	410
August 09, 1951	34	15300
August 13, 1951	22	9800
August 24, 1951	6.0	2400
September 06, 1951	5.1	1960
September 10, 1951	16	7000
September 15, 1951	65	29800
September 18, 1951	27	12100

It is seen from Table 19, that our estimates of daily release on the basis of specific activity of water at the point of complete mixture varied in the range from 410 to 46,000 Ci day^{-1} . Releases occurred on July 5, August 9, September 15 and 18, exceeding 10 kCi can be considered as accidental releases. Taking into account that data from Table 19 are generally comparable with the results of direct measurements (Table 16) we analyzed data from Tables 16 and 19 together in order to reconstruct releases in July-September 1951 and to estimate the ratio between regular and accidental releases. The results of the analysis are presented in Tables 20 and 21.

Table 20. Estimates of daily release of radionuclides from Facility B in July-October 1951.

Month	Number of monitoring days	Regular-to-accidental-releases ratio	Daily-averaged release, kCi day^{-1}	
			Regular	Accidental
July	4	3/1	4.4	42
August	5	4/1	4.0	11
September	10	5/5	5.7	40
October ¹	10	7/3	3.1	190
Average	-	-	4.3	-

Note: daily data for October 1-7, 1951 from Alexandrov et al. (1951) were used; daily-averaged release for October 8-10, 1951 was evaluated with the use of data for the first 10-day period of October from Demyanovich et al. (1952b).

It is seen from Table 20, that monitoring of the releases into the Techa River in July-August 1951 was conducted for 4–5 times a month. For September, there are results of measurements of water samples for the period of September 6–18 and the results of daily monitoring that commenced on September 26. Daily-averaged activity of regular releases in October somewhat decreased (Table 20) correlated with the dynamics of Facility B capacity in this time frame. Daily-averaged regular release for the whole period from July to October 1951 according to our estimates was 4.3 kCi day^{-1} which is in complete agreement with the estimate from Ilyin (1956).

Table 20 shows that cases of high releases, considered as accidental, became more frequent in autumn compared to those in summer. It follows from Table 20, that in Summer 1951, high releases could occur 7-10 times a month and in the Autumn 1951—9-15 times a month. However, taking into account that monitoring was performed only a few days, these values could be overestimated. According to Ilyin (1956), “wild overflows” occurred 4-5 times a month in the beginning of Autumn 1951. Analysis of the monitoring data from the period from the end of 1951 to the beginning of 1952 from Mokrov (2005) provides estimate of the frequency of accidental releases to be 1–3 times a month. It seems, that for evaluation of the total activity released into the Techa River, it could be assumed that accidental releases occurred 1-5 times a month in Summer and 5-7 times a month in Autumn 1951.

Daily-averaged activity of the accidental releases sharply increased in October 1951. As it was discussed before, this could be due to continuous corrosion that aggravated defect of the tanks in Complexes C. This could also be determined by seasonal increase in the level of groundwaters. Data from Tables 16, 19 and 20 provide that averaged value of the accidental releases in the Summer and in the beginning of Autumn 1951 constituted $30 (10\text{--}40) \text{ kCi day}^{-1}$, increased up to $55 (30\text{--}80) \text{ kCi day}^{-1}$ by the end of the September and reached maximum value of 190 kCi day^{-1} in the October.

Table 21 contains estimates of the activity entered the Techa River in July-October 1951 with regular and accidental releases from Facility B, obtained with the use of accepted assumptions and the results of monitoring.

Table 21. Estimates of monthly releases of the radionuclides from Facility B in July-October 1951.

Month	Regular monthly release, kCi month^{-1}	Registered accidental release, kCi month^{-1}	Assumed accidental release, kCi month^{-1}	Assumed total release, kCi month^{-1}
July	136	42	60 (10 – 200)	196 (146 – 336)
August	123	11	60 (10 – 200)	183 (133 – 323)
September ¹	172	198	220 (200 – 260)	392 (370 – 432)
October ²	95	1194	1194	1289
Total	526	1451	1534 (1400 – 1859)	2060 (1940 – 2380)

Notes:

1 – accidental release was evaluated as the sum of registered release and 1–2 cases of possible high releases that could be omitted before September, 25.

2 – accidental release was evaluated as the difference between the total activity released in October 1951 derived from monitoring data (Demyanovich et al. 1952b) and monthly regular releases derived from data in Table 20.

As can be seen from Table 21, during 4 months of 1951 the regular releases into The Techa River constituted about 500 kCi and accidental releases were about 1,500 kCi. According to our estimates, averaged regular-to-accidental-releases ratio in Summer 1951 was 2-to-1. The situation

greatly worsen in Autumn 1951, when the amount of the activity released with cooling waters from Complexes C sharply increased and the regular-to-accidental-releases ratio in October 1951 amounted 1-to-20. It can be stated that the radionuclide composition during this period was completely determined by the age of HLW from Complex C-4. Since Complex C-4 was filled from December 1949, the age of FP in October 1951 did not exceed two years (taking into account one-month storage in cooling ponds of Facility A in December 1949). Radionuclide composition of the releases was to approximately correspond to theoretical mixture of FP. The age of the theoretical mixture of FP can be evaluated using calculated ratios between gamma- and beta-emitters (Gusev et al. 1974). The ratio of “gamma-activity” (in g-equ Ra) to “beta-activity” (in Ci) derived from measurements (Demyanovich et al. 1952b) for October 1951 is equal to 0.09. This ratio is close to the value of 0.11 evaluated for theoretical mixture of FP in conditions of irradiation in reactor for 120 days and storage for 1 year (Gusev et al. 1974).

It is seen from Table 21, that our estimates of the total amount of the activity released into the Techa River in July-October 1951, obtained with use of monitoring data on releases and contamination of the river water in the Techa region upstream from dam D-4 constitutes two million of Ci. Estimation of the activity released into the Techa River before monitoring commenced in July 1951 is possible on the basis of additional analysis of data on the Techa River contamination at the site of releases in Summer 1950 and quantitative analysis of the results of investigations for particular released products and changes in technological processed in LRW management in 1949-1951.

4.2. Comparison of contamination of the bottom sediments in the region of the site of releases in summer 1950 and 1951 and evaluation of the releases in the period from January 1949 to August 1950

The first investigation of the radioactive contamination of the river water and bottom sediments in the Techa River region from the site of releases to Metlinsky Pond was implemented in June 1950 (Ratner et al. 1950; Perminov et al. 1950). Specific activity of the river water and bottoms sediments (BS) was evaluated from gamma-activity of samples. The results of investigations of the river water samples showed that “despite active decantate was conveyed to industrial waters of Facility B, absolute values of specific activity in releases was nearly zero due to dilution.” The results of investigations of BS are presented in Table 22. The results of measurements of the BS sampled in the same Techa River region on August 8, 1951 are given in Table 22 for comparison purposes (Alexandrov et al. 1951).

Table 22. Specific activity of the Techa River bottom sediments sampled downstream from the site of radioactive releases from Facility B in June 1950 and August 1951.

Distance of the sampling points from dam D-2	Specific activity of BS	
	Measurements of June 1950, mg-equ Ra kg ⁻¹	Measurements of August 8, 1951, mCi kg ⁻¹
0.3 km (the site of releases)	5.9	20.6
0.5 km	4.1	-
1.8 – 2.0 km	1.0	18.0
3.1 – 3.5 km	0.3	20.0
5 km (Metlinsky Pond)	background	2.0±1.5

Specific activity of BS can be considered as a cumulated value characterizing releases for the whole preceding period (with account for radioactive decay). It is evident that despite differences in measurement methods (gamma-radiation was measured in 1950 and beta-radiation was measured in 1951), data from Table 22 convince that massive contamination of the Techa River could occur only after June 1950. Analysis of the time-dependence in capacity of Facility B and technological processes for LRW management in 1950 shows that releases into the Techa River were to sharply increase in Autumn 1950 (after repair at Facility B that took place in August 1950) due to the following factors: three-fold increase in plutonium production; onset of releases of the condensate from the diffuser of the main stack (CDMS) and of decantates from Department 3 after their settling in the chromate hole (product 922). Taking all these factors into consideration, it can be supposed that massive contamination of the Techa River commenced not earlier than in September 1950.

This conclusion somewhat contradicts to the estimates of Ilyin (1956) who considered March 1950 as the beginning of the massive releases into the Techa River. However, according to Alexandrov et al. (1951), there were alterations in Department 16 commenced in March 1950 (building of the chromate hole) necessary for reprocessing of decantate from Department 3. According to Ratner et al. (1950) and Perminov et al. (1950), the chromate hole was put into operation in the end of April 1950 and it was filtering until (at the least) July 1950, thus, product 922 did not occur in the Techa River (subsection 2.3.3). In August 1950 facility B was closed for repairs and plutonium was not produced that month (Fig. 2). Thus, only low-level LRW entered the Techa River until the beginning of September 1950; the amount of the low-level LRW can be estimated with the use of data of Ratner et al. (1950), Perminov et al. (1950), Ilyin (1956).

Perminov et al. (1950) estimated the total amount of activity released from Facility B from the beginning of its operation to July 1950 to be equal to 30–35 kg-equ Ra. According to Ratner et al. (1950) and Perminov et al. (1950), only decantates from Department 7 were released into the Techa River until July 1950; moreover, till January 1950 the solutions were purified through iron ore in Department 16 which led to decrease in their activity by a factor of 8-10. It follows from these data, that daily-averaged release into the Techa River in 1949 was 14–16 g-equ Ra day⁻¹ and increased up to 140-160 g-equ Ra day⁻¹ since January 1950. These estimates are comparable with the estimates of Ilyin (1956): 20 g-equ Ra day⁻¹ in 1949 and 300 g-equ Ra day⁻¹ since January 1950. Taking into account that other low-level radioactive wastes were released into the Techa River together with product 757, estimates by Ilyin (which are slightly higher than the estimates by Ratner) are more reliable. Also, taking into account that the contribution of the releases from January 1949 to August 1950 was not significant in the total contamination of the Techa River (follows from data on BS contamination shown in Table 22), estimates of daily-averaged release obtained by Ilyin (1956) could be directly used without revision. Therefore, it can be accepted that the release into the Techa River was 70 Ci day⁻¹ of beta-emitters in January-December 1949 and 860 Ci day⁻¹ of beta-emitters in January-August 1950.

4.3. Evaluation of the releases in the period from September 1950 to June 1951

As it was shown above, levels of regular releases into the Techa River should have been increased in September 1950 as a result of three-fold increase in plutonium production, onset of the release of condensate from the diffuser of the main stack (CDMS) and decantates from Department 3 after their settling in the chromate hole (product 922). The last column of Table 22 shows, that BS in the upper Techa-region had adsorbed significantly large amounts of radioactivity by Summer 1951. This provides evidence for high levels of the releases in precedent period.

Analysis of the history of chromate holes operation (section 2.3) shows that from the Autumn 1950 to the date of launch of the second chromate hole (the date is unknown) product 922 was discharged into the Techa River after settling in the first chromate hole, that became overfull in February 1951. According to our presumptions, uranium production and, correspondingly, release of product 91 (solution of manganous pulp in bisulphate) commenced in the end of 1950. Therefore, there are several factors introducing significant uncertainty into the assessment of the time dependence of regular releases in the period from September 1950 to June 1951. Taking this into account, we cannot assume anything better than to accept a daily-averaged activity of regular releases for this period to be the same as for the following period (section 4.1). According to the assumption, regular releases in the period from September 1950 to June 1951 were equal to 4.3 ± 1.1 kCi day⁻¹ as well as in the period of July-October 1951 (Table 20).

Analysis of the history of operation of Complexes C (section 2.3) allows suggestion that onset of accidental releases into the Techa River commenced during the same period (September 1950 – June 1951). The most probable date for the beginning of accidental releases is April 1951, when there was an extraordinary flood in the Urals as a result of intensive snow melting. Fig. 7 shows seasonal changes in the Techa River flow-rate in 1949-1952.

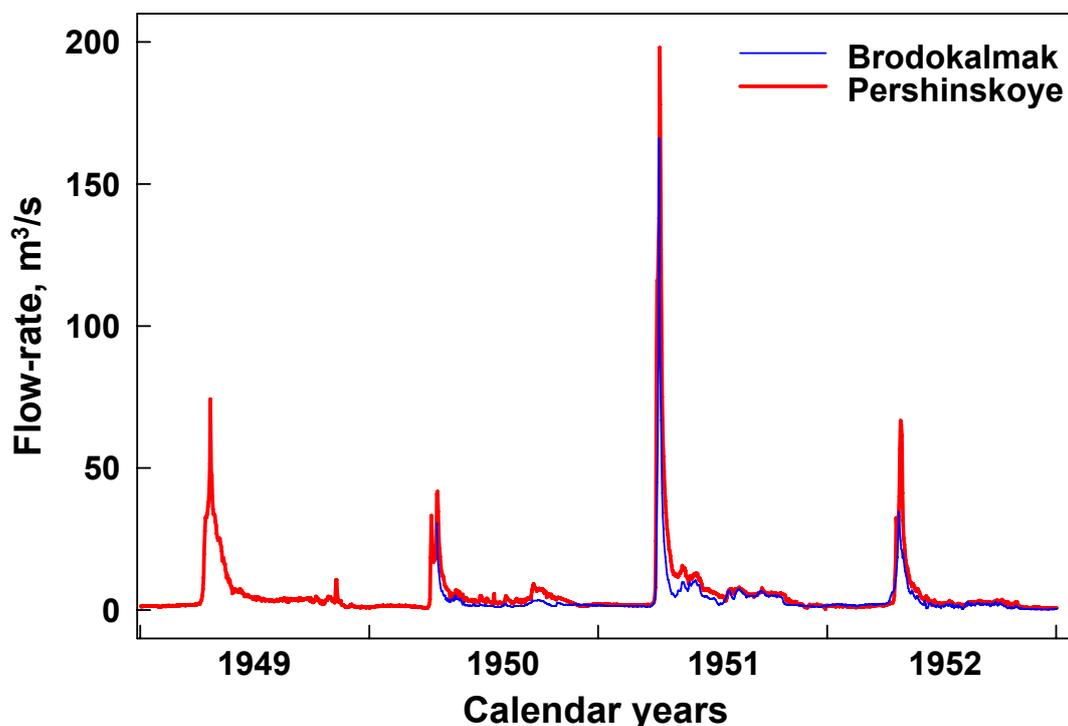


Fig. 7 Seasonal changes in the Techa River flow-rate in 1949-1952.

As can be seen from Fig. 7, the flood of 1951 was much more significant compared to the others. It is evident, that the flood of 1951 caused a sharp increase in groundwater levels resulted in floating of the storage tanks in Complexes C and their damage. The absence of monitoring data greatly contributes to the uncertainty in the assessment of the dynamics of accidental releases in the period from September 1950 to June 1951. Thus, as with the regular releases, we accept the activity and the frequency of accidental releases since April 1951 to be the same as for July-August 1951 (Table 21).

4.4. Reconstruction of the dynamics of the total radioactive releases in 1949-1956

Fig. 8 shows the time dependence in the total releases of radioactive wastes from Facility B into the Techa River in the period of 1949-1956, evaluated with the use of data from the reports of Ratner et al. 1950, Perminov et al. 1950, Alexandrov et al. 1951, Demyanovich et al. 1952b, Ilyin 1952 and our assumptions described in previous sections. The dynamics of the releases obtained earlier from the data of Ilyin (1956) and Marey (1959) and used in the Techa Dosimetry System TRDS-2000 for reconstruction of exposure doses for residents of the Techa River villages (Degteva et al. 2000) is shown in Fig. 8 for comparison purposes. As can be seen from Fig. 8, analysis of additional sources of data, found in MPA archives, allowed more precise evaluation of the releases in the second half of 1951 and of the onset of the massive radioactive contamination of the Techa River (September 1950).

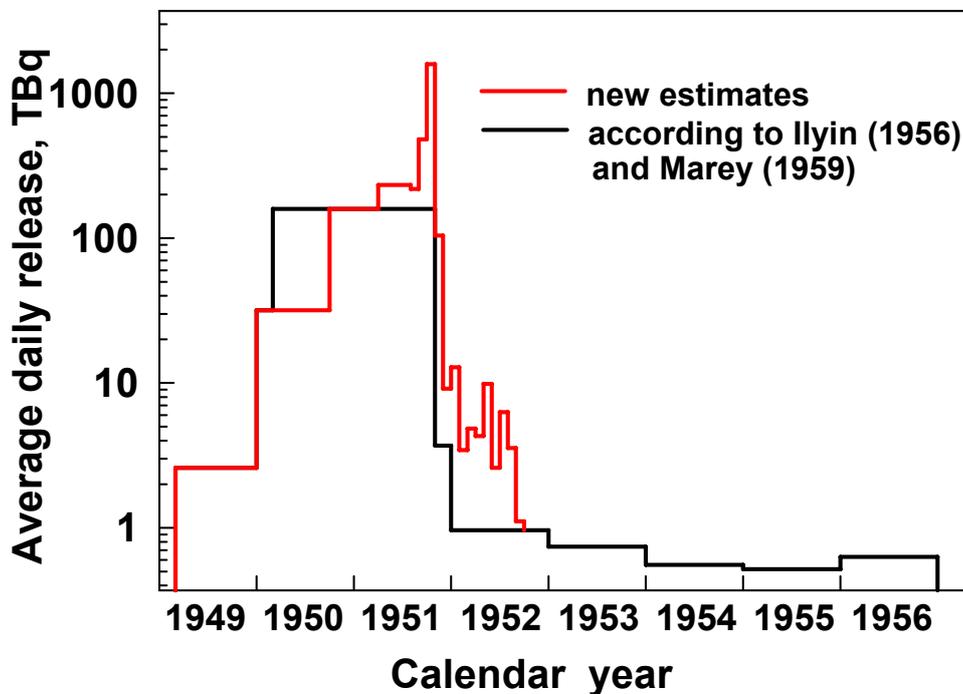


Fig.8. Previous and new estimates of the dynamics of the total activity of radioactive releases from Facility B into the Techa River in 1949-1956.

Comparison of the estimates obtained by Ilyin (1956) with the data from the reports of 1950 (Ratner et al. 1950; Perminov et al. 1950) also allowed confirmation of the daily-averaged release in 1949 and in the beginning of 1950. Estimates of the activity discharged into the Techa River with regular and accidental releases obtained for different times periods are provided in Table 23.

Table 23. Estimates of monthly-averaged releases of the radionuclides from Facility B in 1949-1951.

Time period	Regular releases, kCi month ⁻¹	Accidental releases, kCi month ⁻¹	Total releases, kCi month ⁻¹
January-December, 1949	2.1	-	2.1
January-August, 1950	26	-	26
Spetember, 1950 – March, 1951	130	-	130
April-June, 1951	130	60	190
July 1951	136	60	196
August 1951	123	60	183
September 1951	172	220	392
October 1951	95	1,194	1,289
Total			
(January, 1949 – October, 1951)	2,065	1,720	3,785

It is seen from Table 23, that according to our estimates the total release into the Techa River before it was diverted to Karachay Lake on October 28, 1951, was equal to 2.1 million Ci with regular discharges and about 1.7 million Ci with accidental discharges. Thus, the total release into the Techa River amounts to about 3.8 million Ci (Table 23). This estimate slightly (by a factor of 1.4) exceeds the estimate of Ilyin (1956), who concludes in his Thesis that “Uncontrolled release of radioactive wastes of the plant into the Techa River occurred from 1949 to November 1951. According to approximate assessments obtained for this period, the release into the Techa River of alpha-emitters was 40 Ci, of beta-emitters – 2.73 million Ci and of gamma-emitters – about 1 million g-equ Ra”.

5. DISCUSSION OF THE RESULTS OF THE REVIEW

5.1. Results of archive search

Data and information presented in this report show that the archive search allowed obtaining and analysis of a very important information on the technology for LRW management at Mayak PA during the first years of its operation. Analysis of these data allowed more detailed reconstruction of the history in LRW releases into the Techa River (Appendix). However, a number of questions on the history of the releases could not be completely cleared; for this reason, the Table from Appendix contains approximate estimates that we made from indirect data. It is evident, that more precise reconstruction of the chronology of releases requires further work with MPA archives. For this, it is necessary to continue expert assessment and partial declassification of documents, describing technology for LRW management, containing in CPL archive (dissertation and monograph by M.I. Ermolayev, dissertation by B.A. Zaytsev) and in archive of Mayak PA Administration (Orders and instructions at Facility B, correspondence in the period of 1950-1951). It is also necessary to systematize and continue the work with materials from archive of Plant 25 (Facility B) for the period of 1950-1951 (this archive is still not systematized and, therefore, could not be evaluated for availability of data required for the reconstruction of LRW releases into the Techa River). Archival documents, noted above, can shed light not only on the history of releases but also on methods for radiometry and radiochemical analysis that were used at Mayak PA in the beginning of 1950s and, as a result, will help to make evaluation of the releases more precise.

Nevertheless, it can be stated, that the review of archival documents resulted in great

advance in our understanding of the situation with the release of LRW into the Techa River in 1949-1951. When developing the Techa River Dosimetry System TRDS-2000 we considered the radiochemical facility as a “black box” due to the absence of reliable information. However, the radionuclide composition and the dynamics of the releases in TRDS-2000 is based on expert estimates obtained by D.I. Ilyin (1956), confirmed by the International group of experts in 1997 (JNREG 1997). At present time, the following information became accessible: (1) monthly data on the hold-up time of irradiated uranium blocks in cooling ponds of the reactor plant in 1949-1954 (Glagolenko et al. 2006b); (2) description of the technology for LRW management in 1949-1952 (Ratner et al. 1950; Alexandrov et al. 1951; Demyanovich et al. 1952a; Starik et al. 1952); (3) 10-day period data of the monitoring of LRW release into the Techa River since October 1951 (Demyanovich et al. 1952b); (4) measurements of specific activity and radiochemical analysis of particular released products (Perminov et al. 1950; Alexandrov et al. 1951; Demyanovich et al. 1952a; Starik et al. 1952). These data allows more detailed and precise evaluation of the dynamics and radionuclide composition of the releases for the next version of the Dosimetric System, TRDS-2008.

5.2. Evaluation of methods for radioactivity measurement

Detailed description of the methods for radiometry and radiochemical analysis used at Mayak PA in the beginning of 1950s is only given in dissertation of D.I. Ilyin (1956). We commenced investigation of the possibilities and restrictions of the methods for radiation detection via Monte Carlo modeling of the radiation transport from a source to the detector and absorption in the detector. For gamma-radiation, the geometry of source-detector location, additional filters, type of detector and its calibration were made according to the description provided in Ilyin (1956). However, it is not known if this description corresponds to the methods used at the CPL at earlier times and at the Analytical Laboratory of Facility B. It is evident, that the period of interest characterized with rapid improvement in instrumental methods for radiation measurements. Authors of the reports of 1950 (Perminov et al. 1950; Ratner et al. 1950) mostly used measurements of gamma-radiation for their estimates. Demyanovich et al. (1952b) evaluated radiochemical composition of several samples from gamma- and beta-radiation of particular groups of radionuclides. Alexandrov et al. (1951), Starik et al. (1952) and Ilyin (1956) obtained estimates of radiochemical composition of releases only from measurement of beta-radiation. Measurements of gamma-radiation of different types of LRW are also provided in these reports, however, there were not used for estimates.

Method for detection of gamma-radiation with the lead filter, applied at the CPL (Ilyin 1956) did not allow detection of low-energy gamma-emitters (^{141}Ce and ^{144}Ce). At the same time, in the absence of the lead filter, the results of measurements for these radionuclides could be from 3 to 5 times higher than real values. Method for detection of beta-radiation with end-window counter and mica window of 3-4 mg/cm² (Ilyin 1956) included calibration with a filter that cut off beta-particles with the energy of 0.2 MeV and less. This could result in measurement error at detection of beta-emitters with low energy of electrons (such as ^{103}Ru , ^{95}Nb , ^{95}Zr). Possible systematic errors in radiometry could be one of the reasons that the ratio of gamma-to-beta radiation for several samples did not correspond to theoretical values calculated for mixture of FP of different ages. Investigation of the possibilities and restrictions in the radiometry methods used at Mayak PA in 1950-1956 is underway at present time. When the investigation is completed, correction factors can be evaluated to take account for possible systematic errors in measurements dependent on the energy of beta-particles or photons.

Large uncertainty is associated with measurement of samples, containing significant amount of non-soluble precipitates. Demyanovich et al. (1952b) recommended considering the results of

such measurements as “sufficiently approximate”. It should be noted, that 60-90% of beta activity of the total release concentrated on precipitate and the volume of precipitate was in the range from several hundredths to several tenths of % (Alexandrov et al. 1951). Radiochemical composition of solution and precipitate significantly differed: Transparent solution contained all cesium and about 75% of strontium; while almost all REE were in precipitate (Alexandrov et al. 1951). In these conditions it is possible that in case of insufficient mixing of a daily or a weekly probe, a sample taken for measurement of gamma-radiation and a sample taken from the same probe for measurement of beta-radiation could contain different amount of precipitate. Therefore, samples for measurements of beta- and gamma-activity were not always of completely identical radionuclide composition. This could also be a reason for the fact that some probes were characterized by ratios of gamma-to-beta-activity that did not correspond to theoretical values calculated for mixture of FP of different ages.

It is suggested in Mokrov (2005) to use the ratio of gamma-equivalent of Radium (M) to the total activity (Q) in order to evaluate the reliability of the results of measurement of gamma- and beta-activity of the same sample. It is known that the M/Q ratio for non-separated mixture of FP depends on duration of uranium irradiation in reactor and hold-up time. According to (Gusev et al. 1974), if the irradiation in a reactor is 120 days, the value of M/Q decreases with time from 0.25 (hold-up time is 45 days) to 0.22 (hold-up time is 150 days). As an example, we consider a series of eight measurements of gamma- and beta-radiation of week samples of product 901 from (Demyanovich et al. 1952b) that is characterized with average values of $M=40$ mg-eq Ra/L and $Q=163$ mCi/L with the range of measured values of individual samples of 20-70 mg-eq Ra/L and 64-450 mCi/L. The ratio of averaged values M/Q for this series of measurements is 0.246 and the range of ratios of individual samples is 0.13-0.46. The measurements were performed in 03.03.1952 – 28.04.1952 when the effective hold-up time was equal to 85 days (Glagolenko et al. 2006b), therefore, the theoretical value of $M/Q = 0.234$ is close to the ratio of average values of M and Q in considered series of measurements (0.246).

Table 24 shows the results of stochastic modeling of the M/Q ratio with Monte Carlo method in assumption of normal distribution of measured values with different coefficients of variation. It is seen from Table 24, stochastic estimates of M/Q (equal to 0.24-0.28) are biased (overestimated), and the bias increases with the increase of measurement error. Comparison of the modeling results with the range of M/Q values for measured activity of gamma- and beta-radiation shows that the range of ratios for individual samples (0.13-0.46) is in agreement with the modeling results (0.13-0.48) at the following conditions: measurement error for detection of gamma-radiation is 20% and for beta-radiation is 30%.

Table 24. The results of stochastic modeling of measurements of gamma- and beta-activity of non-separated mixture of FP with the age of 85 days (theoretical value of M/Q is equal to 0.234)

Measurement error %		Stochastic estimate of average ratio M/Q	90% confident interval for M/Q
M (gamma)	Q (beta)		
10	15	0.24	0.18 – 0.33
20	30	0.25	0.13 – 0.48
10	40	0.28	0.13 – 0.63

The results of modeling show that random errors of measurement of gamma- and beta-activity result in bias of M/Q ratio, and the bias from real value for individual samples depends on

measurement error and can be significant. For this reason, we suppose that application of the M/Q ratio as a criteria for reliability of measurements and application of measured values for estimation of real age of FP in the mixture of radionuclides should be performed with caution.

5.3. Assessment of the dynamics and radionuclide composition of releases

It should be noted that all estimates of the total activity provided in this report are based on the results of measurements performed in 1950-1952 without any correction coefficients. When our investigation on evaluation of methods for measurements is completed, the values of the activity of releases can be corrected.

As it was shown in Section 4, analysis of data on monitoring of the releases into the Techa River, found in MPA archives, allowed more precise evaluation of the releases in the second half of 1951. Data from reports issued in June-July 1950 allowed more precise evaluation of the releases in 1949 and the first half of 1950, as well as determination of the beginning of massive contamination of the Techa River (September 1950). Estimates for the period from September 1950 to June 1951 are characterized with large uncertainty since they are dependent on our assumptions on characteristics and amount of releases in this intermediate period, for which it was not successful to find any data. It is evident, that the following stage of our study on evaluation of uncertainties will include all acceptable assumptions on characteristics and amount of the releases in this period.

Despite the uncertainties considered above, data given in Fig. 8 and Table 23, show that estimates of the dynamics of releases significantly changed compared to data from Ilyin (1956) that were used in Dosimetric System TRDS-2000. It was thought earlier that the amount of release from March 1950 to October 1951 was at the same level; however, it is seen from Fig. 8, that this period characterized by significant increase in the releases, reaching the maximum value in the first half of October 1951.

The report by Demyanovich et al. (1952b) is the only source for 10-day periods monitoring data on releases into the Techa River since October 1951. According to these data, a total of 1.2 million of Curies was released into the Techa River over the first two 10-day periods of October 1951. Taking into account, that Alexandrov et al. (1951) and Ilyin (1956) noted “wild overflows” on October 8, 9, 10 and 11, it can be supposed that the most part of this activity was released over these several days. Seemingly, this was a forced release of HLW from “worn-out” tanks of Complex C-4. Sharp changes in water regime in October 1951, considered below, also point to this presumption.

Archival documents indicate, that on October 6, 1951, 75-80% of water from Koksharovsky Pond was discharged into Metlinsky Pond, at the same time, water from Metlinsky Pond was discharged through Dam D-4 over 10 hours with the flow-rate of 15 m³/s (Mishenkov 1951). As a result, after evacuation of 300,000-320,000 m³ of water, the Koksharovsky Pond was “partly freed” for further sharp (“volley”) release of LRW from Facility B that occurred on October, 8-11. Measurements of specific activity of water from Koksharovsky Pond performed on October 9-11, 1951, indicated 90-180 microCi/L; specific activity of water in Metlinsky Pond was 57 microCi/L on October 11 (Alexandrov et al. 1951). After that, “according to a direction of Slavsky [Ed. Note: Director of Mayak PA], Koksharovsky and Metlinsky Ponds were washed off” (Ilyin et al. 1951). From October 17 to October 22, clean water was discharged from Kyzyl-Tash Lake into Koksharovsky Pond with the flow-rate of 30 m³/s and dam beams of Dams D-3 and D-4 were completely open. It is indicated in Ilyin et al. (1951), that “More than 15 million of m³ of water was released over 6 days”. Such washing result in a decrease of specific activity of radionuclides in water, however, significant amounts of contaminated bottom sediments entered into the Techa

River with the water from Koksharovsky and Metlinsky Ponds.

It should be noted, that since August 1951 residents of Metlino were prohibited to use the river water for drinking. As a result, the sharp release of LRW in October 1951 probably did not influence the intake of radionuclides for residents. Also, subsequent washing of the ponds resulted in a decrease of gamma-dose rates in air along their shorelines because contaminated bottom sediments transferred to subjacent areas of the Techa River bed. Concerning residents of settlements located downstream, dilution of the release with clean water from Kyzyl-Tash Lake, seemingly, allowed prevention of radionuclide intakes. However, radioactive sediments washed out from the ponds, settled in subjacent areas of the Techa River bed and resulted in an increase in gamma-dose rate in air along its shorelines. Therefore, the sharp release and subsequent washing of the ponds resulted in an increase of the dose of external exposure for residents of the settlements located downstream from Metlino.

Comparison of data on radiochemical composition of LRW (Tables 3, 5, 8, 10, 12 and 14) with the history of releases into the Techa River (Table A1) shows, that the radionuclide composition of the total release should have changed in time. Since 1949 until Autumn 1950, the composition of the total release was determined by the radionuclide composition of the alkaline decantate from Department 7 (product 757). According to Alexandrov et al. (1951), radioactive strontium contributed about 60% and radioactive ruthenium contributed 35% to the total activity of product 757. From Autumn 1950 to Spring 1951 the main contribution into the total release was due to CDMS, decantate from Department 3 after precipitation of barium chromate and barium carbonate (product 922) and solution of manganous pulp in bisulphite (products 91 and 92). The mixture of these types of LRW contained the five groups of radionuclides that could be extracted and measured in the beginning of 1950s (see Section 3.1.3). Seemingly, the most appropriate estimate of the radiochemical composition of the total release for this period is the estimate from Alexandrov et al. (1951) shown in Table 15. According to these data, about half of the total activity was due to REE group, about twenty percent of the total activity was contributed by radioactive strontium, the same – by zirconium and niobium and the rest ten percent was contributed by ruthenium and cesium groups.

Since Spring 1951, radionuclide composition of the total release was influenced by accidental leaks of HLW from Complex C-4 and by Autumn 1951 such influence became determinant. Seemingly, the radionuclide composition significantly varied in time at this period: on the days of “wild overflows” the radionuclide composition was determined by the composition of HLW from tanks C and on other days the radionuclide composition was the same as in previous period (from Autumn 1950 to Spring 1951).

As it was discussed before, radionuclide composition of the HLW in tanks C possibly corresponded to theoretical mixture of FP of a definite age. Unfortunately, we could not find data on the filling of tanks in Complex C-4 in the archives. However, some information could be obtained from the results of measurements of water in canyons of five tanks in Complex C-4 filled with HLW (Alexandrov et al. 1951). These results are shown in Table 25.

It is seen from Table 25, that canyons of the five tanks were contaminated with radioactive substances. Maximum specific activity of water was observed on October 9 in the canyon of tank 16 and was equal to 815 mCi/L and on October 6 in the canyon of tank 12 and was equal to 114 mCi/L. Water in the canyon of tank 12 was repeatedly measured on October 8, and the specific activity significantly decreased, indicating a large supply of cooling water into the canyon before this. However, it is evident that the cooling could not change the age of FP in the tank, therefore, the change in the M/Q ratio from 0.22 to 0.15 could only be explained by measurement error. This fact additionally proves the above conclusion that the exact age of mixture of FP cannot be determined from measured M/Q ratios. Nevertheless, data from Table 25 allow making conclusion,

that HLW in tanks 12, 16 and 18 ($M/Q = 0.15-0.2$) were quite old (the tanks, seemingly, were filled in 1950 году), and that filling of tank 20 ($M/Q = 0.32$) commenced not earlier than the middle of 1951.

Table 25. The results of measurements of gamma- and beta-activity of water in canyons of tanks I Complex C-4 filled with HLW (according to Alexandrov et al. 1951).

Number of tank	Date of measurements	M, mg-equ/L	Q, mCi/L	M/Q	Comment
7	04.10.1951	0.2	-	-	Cooling and leaks of HLW on September 29 and October 2, 1951
12	06.10.1951 08.10.1951	25.6 1.3	114 9.1	0.22 0.15	Supposed leak of HLW on October 8, 1951
16	09.10.1951	131	815	0.16	Supposed leak of HLW on October 9, 1951
18	05.10.1951	3.7	17.8	0.21	-
20	09.10.1951	3.8	11.7	0.32	Cooling and leaks of HLW on September 27

Alexandrov et al. (1951) explained “wild overflow” of October 27, 1951, by leaks from tanks 7 and 20, and the increased release of October 2, 1951, – by the leak from tank 7. The M/Q ratios in the released solutions in these days were 0.54 and 0.82, respectively (Table 16). Despite the ratios were higher than a theoretically possible limit for the M/Q ratio, it can be concluded, that tanks 7 and 20 contained fresh HLW.

Regarding the sharp release of October 8-9, it could, seemingly, be explained by leaks from tanks 12 and 16 that contained quite old HLW. This is confirmed by the fact, that the M/Q ratio for releases occurred in the first and the second 10-day periods of 1951 was very small and was equal to 0.07-0.08 according to (Demyanovich et al. 1952b).

Considering the above, it can be concluded, that radionuclide composition of the “wild overflows” of September 27-29 and October 8-11, 1951 differed significantly. It can also be concluded, that the average age of FP in the sharp release of October 8-11, 1951, with the total activity of 1.2 million of Curies, exceeded one year. It is not possible to make a firm conclusion on the radionuclide composition of the accidental releases in the period of Spring-Summer 1951, because monitoring data are absent. However, according to our assumptions made from measured M/Q ratios, tanks 7 and 20 in Complex C-4 were filled since the middle of 1951, therefore, it is possible that leaks of HLW occurred in Spring and Summer 1951 from tanks 12, 16 and 18 that had been already filled up.

For approximate estimate of the total release and its radionuclide composition, D.I. Ilyin (1956) used the results of measurements of specific activity and radiochemical composition of water and bottom sediments of the Techa River up to its entry into Iset' river. From these measurements he evaluated reservoirs of the radionuclides in the river system in 1951 (presented in Table 26) and then he used balance ratios to evaluate the total release.

Table 26. Approximate estimates of radionuclides reservoirs in the Techa River system (kCi) in 1951 according to Ilyin (1956)

Area of deposits	⁸⁹ Sr	⁹⁰ Sr	¹³⁷ Cs	REE	Zr+Nb	Ru	Σ
Koksharovsky and Metlinsky Ponds	210	220	310	670	370	560	2340
Between Metlino and Muslyumovo	89.5	68	34.5	48	-	33	273
Between Muslyumovo and Zatechenskoye	13.8	21.4	5.2	13.7	-	8.9	63
Carried out to Iset' River	2.7	10.3	0.58	4.8	-	5.6	24
Total	316	320	350	737	370	608	2700

It should be noted that the estimates of Ilyin (1956) on reservoirs of ⁹⁰Sr, ¹³⁷Cs, ⁹⁵Zr+⁹⁵Nb and REE are in full agreement with the amount of released activity. However, for ⁸⁹Sr he gives a slightly lower estimate of the released activity (240 kCi) and for ^{103,106}Ru – a slightly higher estimate (710 kCi) compared to the estimates of the reservoirs of these radionuclides in the river system (316 and 608 kCi, respectively). The reason for such differences is not discussed by the author. Nevertheless, it seems evident, that D.I. Ilyin tried to evaluate the amount and radiochemical composition of the total release of radionuclides into the Techa River with technological and accidental releases from Mayak PA. The composition of the total release in 1949-1951 according to estimates of Ilyin (1956) is presented in Table 27.

Table 27. Radiochemical composition of the total release from Mayak PA into the Techa River in 1949-1951 according to estimates of Ilyin (1956). Contributions of groups of radionuclides are given as percentages.

⁸⁹ Sr	⁹⁰ Sr	¹³⁷ Cs	REE	⁹⁵ Zr+ ⁹⁵ Nb	^{103,106} Ru
9 – 12	11 – 12	12 – 13	26 – 27	13 – 14	23 – 26

It should be noted, that this radiochemical composition was given in Ilyin (1956) as an averaged composition for the period from March 1950 to October 1951, when over 95% of the total activity of 1949-1956 was released into the Techa River. As a result, sharp releases of September-October 1951 were “smoothed” (“smeared over time”) in the estimates of Ilyin (1956) by averaging with much lower levels of releases occurred in earlier times (especially in Spring 1950). Due to the “smoothing”, the ratio of ⁸⁹Sr-to-⁹⁰Sr in the total release became very close to the value character to the mean age of FP of one year. Taking into account that such approach averages radiochemical composition of “young FP” (with the age of 60-90 days) with radiochemical composition of sharp releases of October 1951 (with the age of FP exceeding one year), it cannot be excluded that estimate of the contribution of short-lived radionuclides into the total release evaluated by Ilyin (1956) was not far from the reality.

6. CONCLUSIONS

A significant number of document concerning releases from Mayak PA into the Techa River in 1949-1956 were found as a result of archival search. The following data were analyzed under the project: Monthly data on hold-up time of irradiated uranium blocks in cooling ponds of reactor

plant in 1949-1954; features in the technology for LRW management in 1949-1952; 10-day periods data on monitoring of LRW releases into the Techa River since October 1951; methods and the results of measurements of specific activity and radiochemical analysis of particular released products and total release. This resulted in great advance in our understanding of the situation with the release of LRW into the Techa River in 1949-1952.

Methods for measurement of gamma- and beta-radiation, methods of radiochemical analysis used at Mayak PA in the beginning of 1950s were analyzed. Methods for radiation detection were investigated for possible systematic errors due to different photon energies. Such investigation was conducted with the use of Monte Carlo modeling of radiation transport from a source to a detector and radiation absorption in the detector. Investigation of the possibilities and restrictions in the radiometry methods used at Mayak PA in 1950-1956 will allow evaluation of correction factors to take account for possible systematic errors in measurements dependent on the energy of beta-particles or photons.

Analysis of additional sources of data found in Mayak PA archives allowed more precise estimate of the amount of releases in the second half of 1951 and more precise evaluation of the beginning of massive contamination of the Techa River that occurred in September 1950. Estimates of the time-dependency in the releases significantly changes compared to those used in dosimetric system TRDS-2000. It was previously thought, that the amount of releases was at the same level from March 1950 to October 1951, however, it is evident now that this period characterized by a significant increase of discharges reaching the maximum value in the first half of October 1951, not long before the main technological lines were routed into Karachay Lake.

Comparison of data on radiochemical composition of different types of LRW with the history of releases into the Techa River showed that the radionuclide composition of releases substantially changed in time. The radionuclide composition of the total release was determined by the types of LRW dominating at certain periods of time. Particularly, it was shown that the sharp release of October 1951, when 1.3 millions of Curies were released over one month, was determined by leaks of old high-active wastes from the storage tanks of Complex C. Time-dependent changes in radionuclide composition are planned to be taken into account in the reconstruction of doses of population exposure.

It should be noted in conclusion, that estimates obtained in this report cannot be considered as the only possible estimates. There are still gaps in our knowledge on contamination of the Techa River, in spite of the fact that a large number of archival documents and data were found and analyzed. These gaps can only be made up by subjective assumptions and modeling. Assumptions made in this report to make up for a deficiency in data cannot be considered as the only possible. For this reason, the study of uncertainties due to incomplete information on the Techa River source-term should be continued.

It should also be noted, that the amount and radionuclide composition of the releases cannot be directly related with the levels of radionuclides intake with the river water for residents of the Techa riverside settlements. Flow-rate in the river changed over the period of releases resulting in dilution of released solutions. Sedimentation of suspended particles and sorption of radionuclides by bottom sediments occurred in the upper-Techa River region resulting in changes in the radionuclide composition. It can be supposed for intake of radionuclides by the population, that creation of Koksharovskiy Pond in August 1951 had a more significant influence than the sharp release in October 1951. For this reason, continuation of modeling of radionuclide migration in the Techa River system with the account for more precise data on releases and changes in water balance in the ponds and the river is highly important.

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Appendix

**History of the releases of main types of LRW from Facility B into
the Techa River in 1949-1952**

Table A1. History of the releases of main types of LRW from Facility B into the Techa River in 1949-1952

Released product	Period of the release into the Techa River		Comments
	Start	End	
Alkaline decantate from Department 7 (technological product 757)	January 1949	October 28, 1951	Was purified by sorption (on iron ore) before the release until the end of 1949; since January 1950 released without purification
Decantate from Department 3 after precipitation of barium chromate and barium carbonate (technological product 922)	September-October 1950	April-May 1951	Released into the river after settling in the first chromate hole from Autumn 1950 until the launch of the second chromate hole.
	September 17, 1951	October 28, 1951	Released into the river after settling in the first chromate hole since September 17, 1951
Aluminate waters (technological product 673)	December 20, 1950	October 28, 1951	Reprocessed in the chromate hole until December 1950; released without purification since December 20, 1950
Solution of manganous pulp in bisulphite (technological product 91/92)	December 1950*	October 28, 1951	Onset of the release corresponds to the launch of Department 15. Released into the river without purification
Condensate from the diffuser of the main stack (this type of releases was not foreseen in the initial design)	September 1950*	November 1951*	Onset of the release after building of the steel pipe in August 1950. Released into the river without purification
Desorption waters from apparatus and canyons (desorption purification of apparatus and canyons was not foreseen in the initial design)	September 1950*	Middle of 1952	These LRW contained notable amounts of U and Pu and were to be conveyed to tanks C (Demyanovich et al. 1952b). However, "there were cases of the release into the river without preliminary analysis" (Alexandrov et al. 1951). Continued released into the River in April 1952 (Starik et al 1952).
Accidental leaks of high-active wastes from Complexes C	April 1951*	October 28, 1951	Onset of releases is presumably related to extraordinary flood in 1951 Cases of accidental releases into the river were registered after October 28, 1951

* approximate estimates from indirect data