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Artificial Radionuclides in the System: Water, Irrigated Soils, and Agricultural Plants of the Crimea Region

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Abstract: In the frame of the radioecological monitoring after the Chernobyl nuclear power plant accident, the features of migration and distribution of artificial radionuclides in the North Crimean Canal (NCC) irrigation system were studied. Standard methods of radiochemical analyzes and modern radiospectrometric equipment were used. It was determined that the irrigation system of the NCC retains 43–59% ^{90}Sr, 59–60% $^{239+240}$Pu, and 66–70% ^{137}Cs of the concentration radionuclides entering to irrigated fields with the Dnieper waters. The NCC irrigation system plays the role of a buffer against the radionuclide pollution of the Karkinitsky Bay (the Black Sea). Differences in the accumulation of radionuclides by agricultural crops were revealed. The ^{90}Sr and $^{239+240}$Pu transfer factors (TF) for alfalfa were n $\times$ 10^{-2} and n $\times$ 10^{-1}, respectively. The TF for wheat, corn, and rice for ^{90}Sr were n $\times$ 10^{-3}, and for $^{239+240}$Pu—n $\times$ 10^{-2}. A radioecological assessment on the safety agriculture along the NCC was made: in the absence of an increase in the entry of the Chernobyl origin radionuclides with the Dnieper river waters to the NCC, the levels of activity concentration of artificial radionuclides in cultivated crops will not exceed the maximum permissible concentration for food raw materials in the coming years.

Keywords: ^{90}Sr; ^{137}Cs; $^{239+240}$Pu; the Chernobyl NPP accident; the North Crimean Canal; irrigated agriculture; soil; plants; distribution coefficients; transfer factor

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1. Introduction

On 26 April 1986, an accident occurred at the Chernobyl nuclear power plant (ChNPP), which was the largest nuclear disaster of the 20th century [1–3]. During the 10 days, while emissions into the atmosphere were occurring, 1.9 EBq of radioactive material, represented by fission products and transuranic activation products, entered the environment, which amounted to 3–4% of the activity contained in the reactor core. In addition to the inert gases, 20% of the iodine present in the core (670 PBq ^{131}I); 10% of the total caesium (19 PBq ^{134}Cs and 37 PBq ^{137}Cs); 8 PBq ^{90}Sr; 0.1 PBq plutonium alpha-emitting isotopes; and 5.2 PBq of beta-emitting ^{241}Pu, the parent radionuclide for ^{241}Am, were released into the atmosphere [3–5]. The significance of the releases of ^{137}Cs and ^{90}Sr into the environment as a result of the Chernobyl disaster (89 and 7.4 PBq, respectively) can be compared with the release of these radionuclides from the consequences of the use of weapons in open environments: 1300–1500 and 650–1300 PBq, respectively, as well as with the releases of these radionuclides as a result of other nuclear incidents [4,5]. Radioactive contamination of aquatic ecosystems located both near the site of the accident and at a considerable distance from it is associated with the release into the atmosphere and wind transport of radioactive products and aerosol particles. In the first months after the ChNPP accident, the Black Sea was subjected to acute radioactive contamination. In May 1986, 1.7–2.4 PBq ^{137}Cs and 0.3 PBq ^{90}Sr fell onto the surface of the Black Sea. In the post-accident years, the radioecological situation in Crimea was determined by secondary contamination of radionuclides, primarily ^{90}Sr, with river runoff from the Dnieper. It was a chronic radioactive contamination, mainly due to the Dnieper water use from the

North Crimean Canal (NCC) [6–9]. So, 23 TBq of ^{137}Cs and 160 TBq of ^{90}Sr were carried into the Black Sea with the waters of the Danube and the Dnieper rivers from 1986 to 2000 [4,8]. Moreover, the work on the direct decommissioning of the Chernobyl cooling pond has been beginning since January 2010 [10]. These works cause the unavoidable entry of dissolved radionuclides sequentially from the water of the Chernobyl cooling pond into the Pripyat and the Dnieper rivers, then through the cascade of the Dnieper reservoirs, as well through the NCC into the Black Sea ecosystem, inland reservoirs, and irrigated soils of Crimea [11,12].

The NCC was built between 1961 and 1971 to assure a sustainable water supply to the south of Soviet Ukraine and Crimea. The NCC originates in the lower reaches of the Kakhovskoye reservoir, the last one in a cascade of six artificial reservoirs built along the riverbed of the Dnieper [11,12]. The width of the canal at its starting point reaches 150 m, and the depth is 7 m. The total length of the main canal of the NCC from the Kakhovskoye reservoir to the city of Kerch in Crimea is 403 km, which makes it the longest canal in Europe. The average annual flow of the NCC reaches 380 $m^3 \cdot s^{-1}$, the maximum is up to 500 $m^3 \cdot s^{-1}$. This is approximately 30% of the total flow of the Dnieper in its lower reaches [11,12]. About 60–80 $m^3 \cdot s^{-1}$of this volume was used for agricultural needs in the southwestern regions of the Kherson region of Ukraine, and another 300–320 $m^3 \cdot s^{-1}$ was delivered to Crimea. The North Crimean Canal was built in the steppe continental zone of Crimea, where additional sources of water were required for irrigated agriculture. The water system of the NCC includes the main canal and irrigation canals, through which the Dnieper water enters the agricultural land, as well as outlet canals, through which the water used for irrigation is discharged. The main canal of the NCC goes east to the city of Kerch. The branched western part of the canals was used mainly for irrigation of agricultural land in the northwestern part of Crimea, the total area of which was more than 400 thousand hectares until 2014 [9,13]. In general, about 80% of the water supplied through the NCC was used for the needs of agriculture in Crimea, including 60% that was used to assure the cultivation of rice. In the southwestern part of the Kherson region of Ukraine and the northwestern regions of Crimea, where the main areas of irrigated fields were located, water was discharged into the Karkinitsky Bay of the Black Sea.

The radioecological studies of the aquatic ecosystem of the NCC were carried out periodically until 1991 [14]. In 1991–1992, the comprehensive radioecological monitoring was organized on the basis of the A.O. Kovalevsky Institute of Biology of the Southern Seas (IBSS) of NAS of Ukraine within the framework of the joint international program: "Program of Urgent Measures to Eliminate the Consequences of the ChNPP Accident" (the IBSS and ENEA-DISP (Rome, Italy) [15]. Studies were carried out, the purpose of which was to assess the contribution of the NCC irrigation system to the migration of the Chernobyl ^{90}Sr, ^{137}Cs, and transuranic elements from the Dnieper water to the irrigated soils of Crimea's cultivated plants. In 1995, the radioecological studies in the system of water of the NCC–irrigated soils–irrigated plants were completed due to the lack of funding on state themes. For the period of the studies, the important role of the aquatic ecosystem of the NCC was revealed as a factor in the inflow of secondary radionuclide contamination from the ChNPP accident area to the irrigated lands in the south of Ukraine and Crimea [9,15–21]. From 2014 to 2022, the NCC did not function.

It is known [9,14,22,23] that until 2014, the Dnieper water, which came from the accident area to Crimea through the NCC, was a factor of the chronic secondary radioactive contamination by radionuclides of ^{90}Sr, ^{137}Cs, and isotopes of plutonium and americium of the Black Sea waters, the inland waters of Crimea, and the vast irrigated territories of the Crimean Peninsula. Therefore, the study of the role of the NCC in the transport of radionuclides ^{90}Sr, ^{137}Cs, plutonium, and americium with the Dnieper river water is of particular relevance in the contemporary period.

In 2022, after the resumption of the entry of the Dnieper water to the NCC, the samples of bottom sediments, water, and suspended matter in the main canal near the region of Armyansk and Krasnoperekopsk, as well soil in the wheat field, were taken. These

data will serve as conditional background values for tracking changes in the levels of the technogenic radionuclide activity concentration and determining trends in changes in the radioecological situation in the canal and irrigated fields of the NCC irrigation system in the future.

The aim of the research is to generalize the results of radioecological monitoring of the features of migration and distribution of the ^{90}Sr, ^{137}Cs, $^{238, 239 + 240}Pu$, and ^{241}Am in the NCC irrigation system; to calculate the quantitative characteristics of the entry of these technogenic radionuclides into the Karkinitsky Bay of the Black Sea after irrigating of the agricultural land; to evaluate the role of the NCC system in the transfer of long-lived radionuclides from the Dnieper water to irrigated soils and cultivated crops in the period after the ChNPP accident; and to perform a radioecological assessment on the safety of conducting agriculture along the NCC for the next years.

2. Materials and Methods

2.1. Sampling Site and Materials

To achieve the goal of the research, an analysis of the literature data and the results of our own long-term studies were carried out. Five main stations along the main canal of the NCC were selected for sampling. Samples of water, suspended matter (water filtration on filters with a pore diameter of 0.45 μm) and bottom sediments were sampled on these stations, as well as on the stations 4A and 5A. Samples of soils, and cultivated crops from adjacent irrigated agricultural lands were collected on the test sites 1 and 2. A map-scheme of sampling stations is shown in Figure 1.

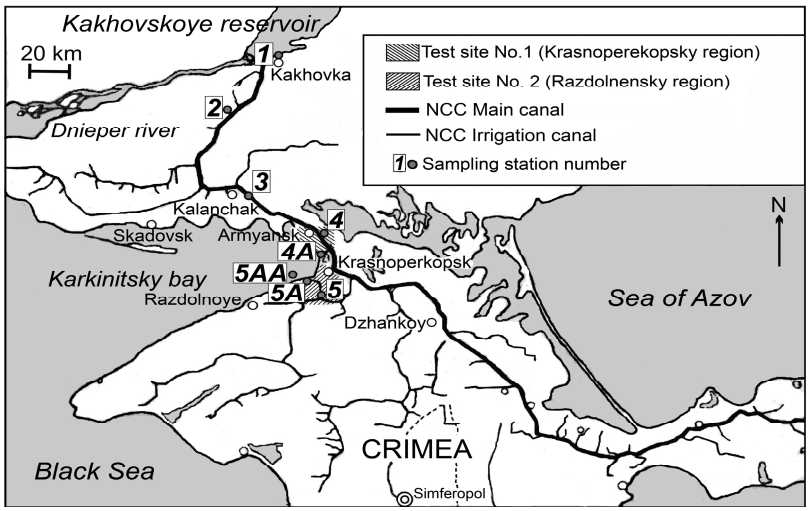

Figure 1. Map-scheme of sampling in the area of the North Crimean Canal.

The sampling stations were located along the NCC: station 1 (0 km, start of the NCC, point whence water is supplied from the Kakhovskoye reservoir) and stations 2–4 (28, 84, and 125 km from the beginning of the canal). Two test sites were selected in two main rice-growing regions of the Crimean Peninsula (Krasnoperekopsky and Razdolnensky) to study the contribution of irrigated agricultural land ecosystems to the extraction of the "Chernobyl" radionuclides from the Dnieper water. The test site 1 was located on the plot between the cities of Armyansk and Krasnoperekopsk (Figure 1). The supply of the Dnieper water to this territory was constantly monitored (station 4). A pumping station (station 4A, 130 km from the beginning of the canal), through which discharge water was pumped into the Karkinitsky Bay of the Black Sea, is situated at the water outlet from this test site (Figure 1). A similar test site 2 was chosen in the Razdolnensky region of Crimea (stations 5 and 5A, 150 and 160 km from the beginning of the canal, respectively) (Figure 1). The land area of the test site 1 is 1380 ha and the area of the test site 2 is 6500 ha. In the study area, there were chestnut and dark chestnut solonetsous soils. Sea water samples

were also taken in the Karkinitsky Bay of the Black Sea (station 5AA, 161 km from the beginning of the NCC). A typical annual operation of supply of the Dnieper water to the irrigated lands of test sites 1 and 2 was as follows: in mid-March—the beginning of supply (0.1–0.4 million m^3), from May to August—the maximum supply (0.7–0.8 million m^3), mid-November—end of water supply to irrigated fields (0.4–0.1 million m^3).

The samples of bottom sediments in the NCC (layer 0–10 cm) were taken with a bottom grab with a capture area of 0.025 m^2. At test sites 1 and 2, soil samples were taken in irrigated fields (surface layer 0–10 cm) by a square frame of 0.05 m^2. On irrigated fields at each station, samples of cultivated plants were taken at three points at each field on an area of 0.25 m^2. At each station, 3 samples of bottom sediments, soils, and cultivated plants were taken, combined into one sample for each object, dried, homogenized, and mixed, and an aliquot was taken for radiochemical analysis for determination of artificial radionuclide concentrations in them.

The following plant species were selected to study the transfer of post-accident ^{90}Sr to agricultural crops grown on irrigated lands of the NCC: alfalfa (*Medicago sativa* L., green mass), wheat (*Triticum durum* Desf. L., stems with an ear), corn (*Zea mays* L., stems, leaves), and rice (*Oriza sativa* L., whole plant, straw, stems with grain).

For carrying out of this research, the following number of samples were selected, processed, measured, and analyzed:

- 205 water samples (including 133 samples for the determination of ^{90}Sr, 48 samples for determination of ^{137}Cs, 24 samples for determination of transuranium elements);
- 45 samples of suspended matter (including 5 samples for the determination of ^{90}Sr, 24 samples for the determination of ^{137}Cs, 16 samples for determination of transuranium elements);
- 96 samples of bottom sediments (including 24 samples for the determination of ^{90}Sr, 48 samples for the determination of ^{137}Cs, 24 samples for determination of transuranium elements);
- 100 samples of irrigated soils (including 26 samples for the determination of ^{90}Sr, 48 samples for the determination of ^{137}Cs, 26 samples for determination of transuranium elements);
- 60 samples of cultivated crops (including 21 samples for the determination of ^{90}Sr, 21 samples for the determination of ^{137}Cs, 18 samples for determination of transuranium elements).

In total, 506 samples were taken for research.

2.2. Methods

2.2.1. ^{137}Cs Procedures

The sorption method of determination of ^{137}Cs radioisotopes in water (50 L is the volume for each sample) is based on the principle of carrying out gamma-spectrometric measuring after pollutant collection on the filters to separate the suspended matter in fibrous or powder sorbents [4]. The suspended matter was collected by filtering large volumes of water (250–1000 L) through a CUNO filtering device (USA), providing separation of suspended matter larger than 0.45 μm. After the ashing of sorbents and filters with particle matter, as well cultivated plants (up to 2 kg dry weight), and drying samples of the bottom sediments and soils (100–150 g dry weight), the samples were measured at the gamma-spectrometer [24]. Measurements of the samples were carried out in NaI(Tl) scintillation detectors (nos. 1, 2) with lead shield, with the ORTEC 855 Dual Spec AMP amplifiers, Canberra AMP2026, and also ultra-pure germanium detector Canberra-PackardXtRa GX2019 at the end with relative efficiency of about 23%. The data were treated according to commonly accepted statistical techniques [24]. The methods used for detecting radionuclides in natural objects have been repeatedly successfully tested within the framework of international intercalibration [4]. The relative error of the ^{137}Cs activity concentration determination was not more than 15%.

2.2.2. ^{90}Sr Radiochemical Procedures

The ^{90}Sr concentration in water was determined in 20 L samples that were taken at the water surface on depth about 1 m. To determine the ^{90}Sr, the samples of bottom sediments (up to 500 g wet weight), soil (up to 200 g dry weight), and agricultural crops (up to 2 kg dry weight) were also taken. Ash aliquots of each sample (100–150 g) were taken for the radiochemical procedure.

The method of ^{90}Sr determination in the environment samples, a standard method in international practice, is based on radiochemical analysis, the measuring of ^{90}Sr activity on its daughter radionuclide ^{90}Y Cerenkov's radiation with using the "low background level" liquid-scintillation counter (LSC) LKB "Quantulus1220", and subsequent mathematical data processing [4,25].

The lower limit of detection (LLD) is 0.01–0.04 Bq·kg^{-1} (or Bq·m^{-3}) of sample. The results are reported as the mean of the values measured for the individual samples/organisms and standard deviation (SD) for each group of data. The stable strontium concentration was determined as the ^{90}Sr chemical yield [4]. The computational scheme, which was used for the determination of the concentrations and errors of method of ^{90}Sr determination in water, bottom sediments, soils, and agricultural plants, allowed for the correct assessment of the degree of contamination by this radionuclide of the investigated objects. The relative error of the received results did not exceed 20%.

The quality control of the analytical methods and the reliability of the calculated results were supported from the constant participation in international intercalibrations during 1990–2004 under the aegis of the IAEA (Vienna, Austria). Results of the IBSS participation in the intercalibration were included in the intercalibration report materials [4] with the following notice: they were accepted as reliable data, as evidenced by a quality certificate, which was received by the author.

2.2.3. $^{238,239+240}$Pu and ^{241}Am Radiochemical Procedures

For the determination of transuranic elements ($^{238,239+240}$Pu and ^{241}Am) in water, 100–500 L of water sample was taken at each station. To obtain 1 sample of suspended matter for the determination of transuranic elements in it, 1400–2000 L of water was filtered. Between 10 and 50 g of ash from each sample was used to determine by radiochemical analysis the transuranic elements in bottom sediments, soils, and agricultural plants.

The determination of the alpha-radionuclides $^{238,239+240}$Pu and ^{241}Am were carried out according to accepted radiochemical techniques [4,25–28]. The procedure is based on thermal and chemical processing of natural samples with the subsequent plutonium and americium adsorption and desorption using ion-exchange resin AG 1 × 2 or Dowex 1 × 2 in the chloride-form with 50–100 and 100–200 mesh size grains or AB–17-8 125–250 μm. After the pretreatment and purification, the Pu and Am radionuclides were electrodeposited on steeliness plates, and the samples were then analyzed in the "EG&G ORTEC OCTETE PC" alpha-spectrometer. The efficiency determination of the detector measuring system and calibration of the energy spectra were completed through standard sources containing radioactive isotope of ^{239}Pu and ^{242}Pu and ^{243}Am [4]. The plutonium alpha-radionuclide ^{242}Pu and ^{243}Am were added to the sample as radio-tracers for the determination chemical yield of Pu and Am, respectively. The data obtained were treated according to commonly accepted statistical techniques [4]. The total error of the ^{239,240}Pu concentration determination did not exceed 13% for samples of bottom sediments, soils, and aquatic organisms, and 30% for ^{239,240}Pu water samples and all samples for ^{241}Am and ^{238}Pu.

2.2.4. Determination of Quantitative Characteristics (Coefficients)

Distribution coefficient of artificial radionuclides by bottom sediments and soils (K_d) was determined by the following ratio [29]:

$$K_d = \frac{A_s}{A_w} \quad (1)$$

where A_s—radionuclide activity concentration in the bottom sediments (soils), Bq·kg^{-1} (d.w.); A_w—radionuclide activity concentration in water of the Dnieper River in the NCC, Bq·kg^{-1}.

For estimating the soil–plant transfer of radionuclides, we used TF (Bq·kg^{-1}/Bq·m^{-2})—the transfer factor [30]:

$$TF = \frac{A_p}{D_R} \quad (2)$$

where A_p—radionuclide activity concentration in a plant, Bq·kg^{-1} (d.w.); D_R—surface activity of a radionuclide in soil (Bq·m^{-2}) on which the plant is grown.

3. Results

3.1. ^{90}Sr in the NCC Ecosystem–Irrigated Soils–Cultivated Plants

The sources of ^{90}Sr entry into the Kakhovskoye reservoir and the NCC (Figure 2) are the atmospheric transport of the radionuclide and its fallout with precipitation immediately after the ChNPP accident, as well as water transport through the cascade of reservoirs of the Dnieper River.

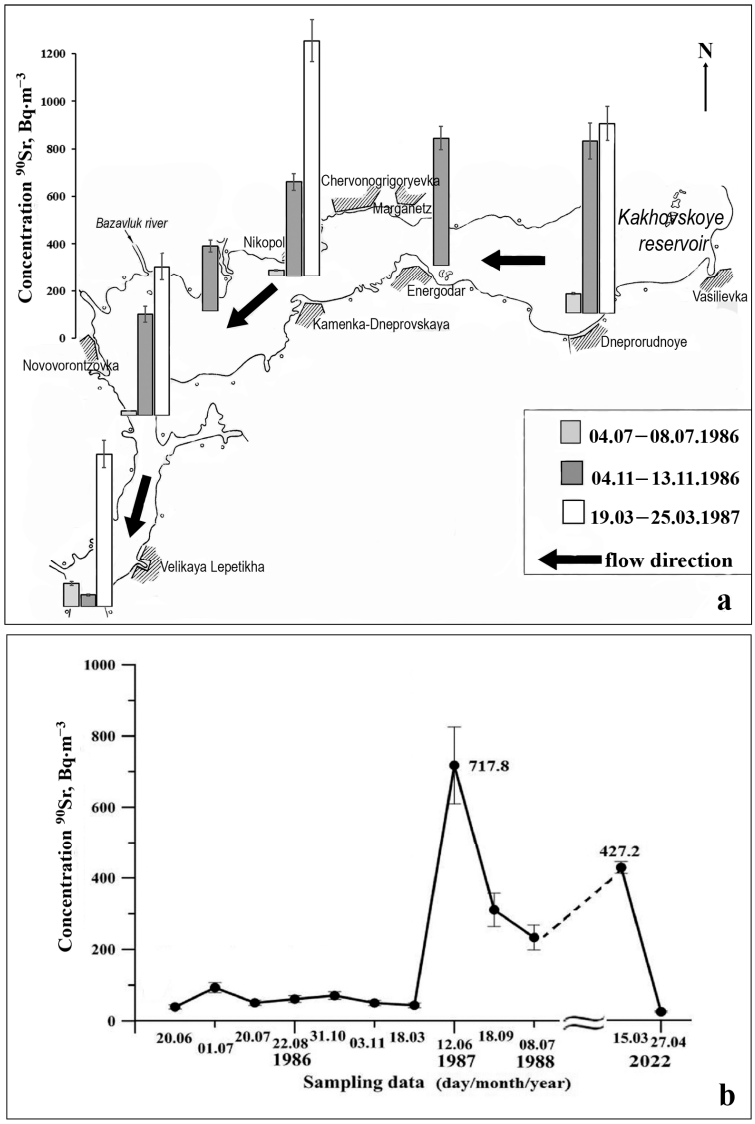

Figure 2. Dynamics of ^{90}Sr concentration in the Dnieper water in the NCC entry to the Crimean Peninsula (Armyansk) in 1986–1988 and 2022 (**b**) compared to ^{90}Sr input into the Kakhovskoye reservoir in 1986–1987 (**a**).

The concentration of ^{90}Sr in the water of the NCC was in the range of 4.1–6.8 Bq·m^{-3} before the ChNPP accident [31]. After the ChNPP accident (07.1986), the concentration of ^{90}Sr in the NCC water near the city of Armyansk (the canal's entry to the Crimean Peninsula) sharply increased (up to 52.3 ± 3.6–61.1 ± 7.3 Bq·m^{-3}) as a result of atmospheric fallout. Significant variations in the concentration of ^{90}Sr along the main canal of the NCC were not observed [32]. The concentration of ^{90}Sr in the water of the NCC increased nine times by June 1987 compared to 1986, which was due to the inflow of water from the upper reaches of the Dnieper into the NCC (Figure 2a). In March–April 2022, the Dnieper water, which had just entered to Crimea after its eight-year absence, was sampled near the city of Armyansk. ^{90}Sr concentrations in water of this station ranged from 427.2 ± 16.4 Bq·m^{-3} in March 2022 to 23.9 ± 2.1 Bq·m^{-3} in April 2022 and corresponded to values of radionuclide concentrations observed at this sampling station in 1986–1987 (Figure 2b). The difference in ^{90}Sr concentrations may have been due to sampling during and after the flood period.

^{90}Sr concentrations in the water of the NCC in 1992–1995 are shown in Figure 3.

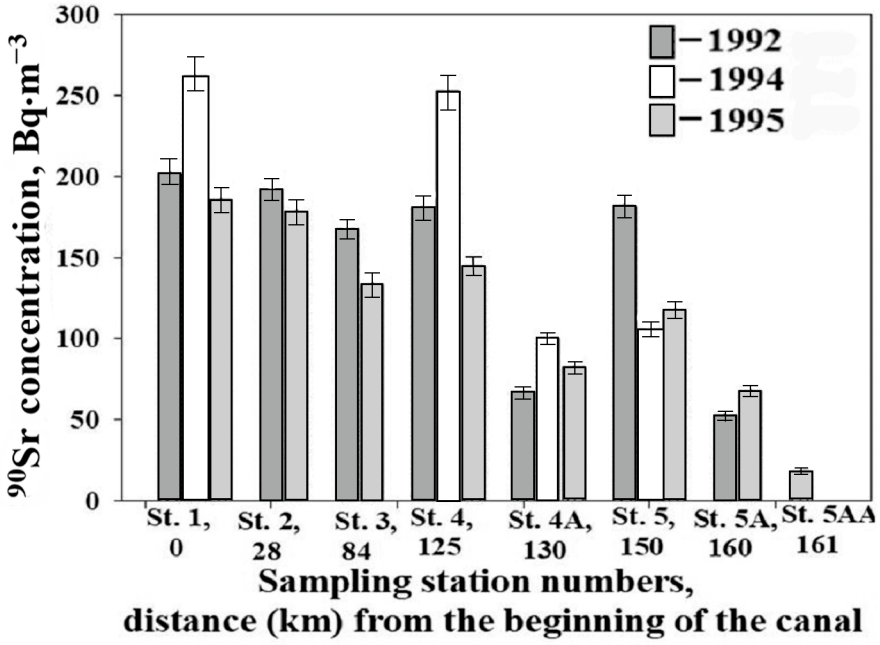

Figure 3. Dynamics of ^{90}Sr concentration in the NCC water in 1992–1995.

The results of research showed that in 1992–1995, in the entry of the NCC (0 km), the concentration of ^{90}Sr was the highest (202.0 ± 9.0 Bq·m^{-3}), and later it was distributed almost evenly at the stations along the main canal of the NCC.

An increase the concentration of ^{90}Sr in 1.4 times in the water of the NCC at stations 1 and 4 in 1994 compared to 1992 was associated with the entry of this radionuclide into the Dnieper with flood waters from the river catchment basin [33].

According to the obtained data, 28.5 GBq of ^{90}Sr is supplied to the irrigated lands: 5 GBq to test site 1 and 23.5 GBq to test site 2.

Taking into account the difference in the values of ^{90}Sr concentrations in the water of irrigation and outlet canals in 1992–1995, it was calculated that 43% and 59% of ^{90}Sr were extracted by ecosystems of irrigated lands in test sites 1 (stations 4 and 4A) and 2 (stations 5 and 5A), respectively (Figures 1 and 3). The amounts of ^{90}Sr after the ecosystem of the irrigated fields in the outlet canals were 57% (test site 1) and 41% (test site 2), respectively.

The volumes of water supply to the canal adopted in 1998 (2095.9 million m^3) [4] were used to calculate the total amount of ^{90}Sr supplied to the irrigated lands of Crimea through the NCC. At the same time, the volumes of water supply for technical needs, to Kerch city and the filling of local reservoirs, were subtracted from the volume of annual water

supply to Crimea through the NCC. In 1986–1999, the total amount of ^{90}Sr brought with the Dnieper water to the territory of Crimea was about 5900 GBq (Table 1).

Table 1. Dynamics of the average annual concentrations (C) of ^{90}Sr in the water of the NCC and the entry of ^{90}Sr into the territory of Crimea in 1986–1999.

Year	NCC Water Volume for Irrigation, Million m^3	C, ^{90}Sr, Bq·m^{-3}	Quantity Measurements/ (References)	Total Activity Brought with Water, GBq
1986	2831.3	59.3	38	167.9
1987	2464.9	522.2	31	1287.2
1988	2480.0	296.0	30	734.1
1989	2585.4	258.5	7	668.4
1990	2961.8	205.0	1	607.2
1992	2842.7	199.0	1	565.7
1993	2842.7	155.0	8	440.6
1994	2842.7	282.1	5	801.9
1995	2842.7	105.6	12	300.2
1999	2003.2	147.5	[33]	309.1
	Entry result:			5882.3

It was noted that regardless of the observation period, the concentration of ^{90}Sr in the water of the NCC was 11–175 times lower than the permissible levels, but 4–63 times higher than the pre-accident levels [34].

There was a total regularity in the distribution of ^{90}Sr concentration in the water and bottom sediments of the main canal of the NCC: the concentration of ^{90}Sr decreased as the sampling stations moved away from the beginning of the NCC (Figure 4).

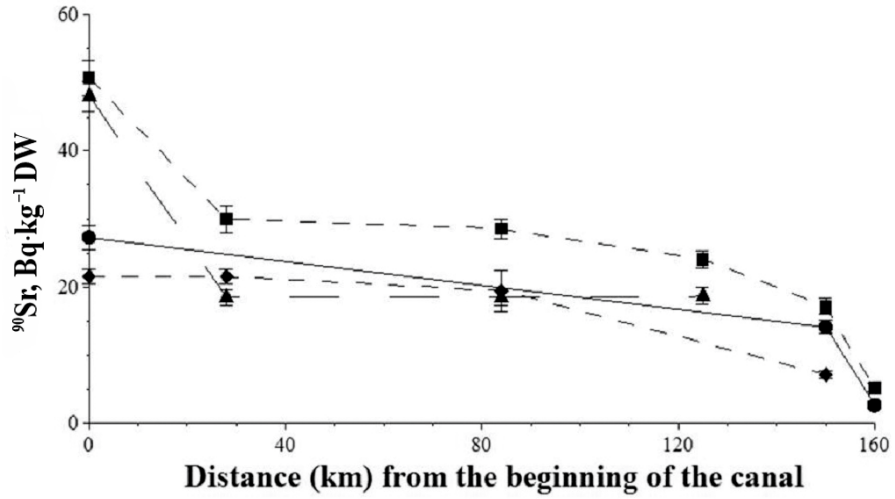

Figure 4. ^{90}Sr concentrations in bottom sediments of the NCC main canal (1992 (●), 1993 (▲), 1994 (♦), and 1995 (■)).

The concentrations of ^{90}Sr in bottom sediments in a distance of 150 km and 160 km of the NCC beginning were 43% and 39%, respectively, of those at the start station of the canal.

An increase in the concentration of ^{90}Sr in the bottom sediments of the NCC from 1992 to 1995 by 1.2–2.6 times was noted, depending on the sampling station (Figure 5).

The results of determining the concentration of ^{90}Sr in the bottom sediments of the irrigation and outlet canals of the NCC (Figure 5) confirmed this observation. The range of ^{90}Sr accumulation coefficients in bottom sediments of the NCC was 51–608 [35].

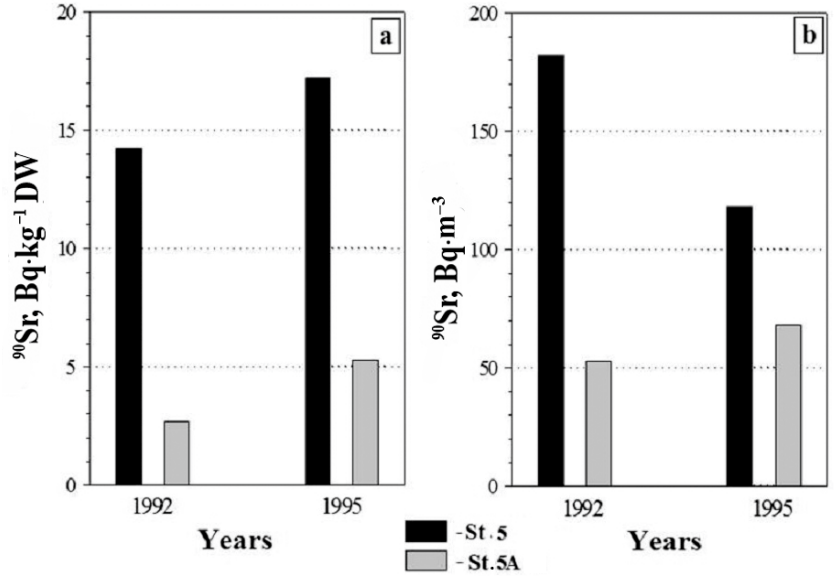

Figure 5. ^{90}Sr concentrations in bottom sediments (**a**) and water (**b**) of the NCC in July 1992 and 1995 in the irrigation (station 5) and outlet (station 5A) canals.

It is obvious that the ^{90}Sr that entered the aquatic environment accumulated in the bottom sediments of the canal, redistributing over time between the abiotic and biotic components of this ecosystem. At the same time, the concentration of ^{90}Sr in water and bottom sediments in the outlet canal was significantly lower than in the main canal, which was probably due to irrigation work along the canal of the NCC (Figure 5).

The highest average concentration of ^{90}Sr (13.5 ± 0.8 Bq·kg^{-1} dry weight) was noted in flooded soils under rice, which is explained by the peculiarity of its cultivation technique (Table 2, Figure 6).

Table 2. Concentrations (C) and distribution coefficients (K_d) of ^{90}Sr in irrigated soils near the NCC area (1992–1995).

Soil	№ Station	Year	Layer, cm	C, ^{90}Sr, Bq·kg^{-1} ± σ, Dry Weight	K_d
Flooded soilunder rice	3	1993	0–10	13.7 ± 0.9	91
	4	1992	0–10	7.5 ± 0.7	41
		1994	0–10	18.6 ± 0.9	74
		1995	0–10	19.1 ± 1.4	132
	5	1993	0–10	12.2 ± 0.3	67
		1994	0–10	11.2 ± 0.7	107
	5A	1995	0–10	12.2 ± 0.8	180
Soil under alfalfa	2	1992	0–10	5.9 ± 0.7	31
		1993	0–5	5.6 ± 0.5	178
		1994	0–5	7.5 ± 0.7	40
		1995	0–10	7.3 ± 0.8	41
	3	1992	0–5	4.4 ± 0.7	26
		1993	0–5	2.5 ± 0.5	17
	4A	1994	0–10	4.1 ± 0.5	41
		1995	0–10	9.2 ± 0.3	112
Soil under corn	2	1995	0–5	7.3 ± 0.8	41
	4	1994	0–5	6.3 ± 0.6	25
	5	1994	0–5	5.1 ± 0.5	48

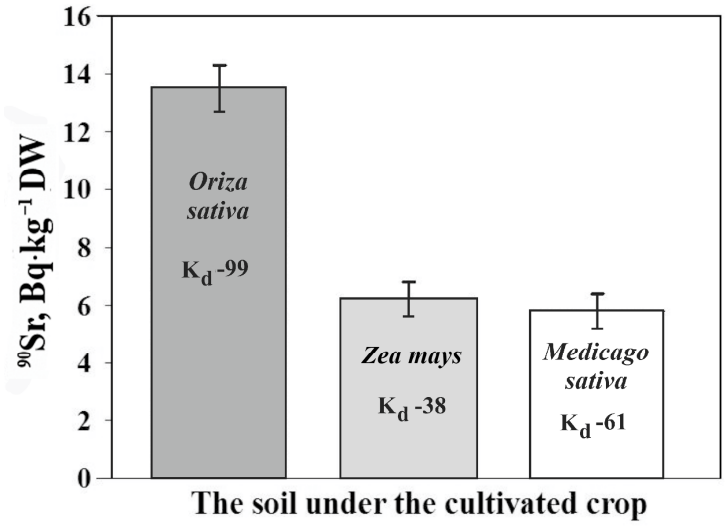

Figure 6. Average concentrations and distribution coefficients (K_d) of ^{90}Sr in irrigated soils under rice, corn, and alfalfa (1992–1995).

At the same time, in the soil under alfalfa and corn, values of ^{90}Sr concentrations were close (Figure 6). The distribution coefficients of ^{90}Sr in soils were calculated for the assessment of the accumulation capacity of ecosystem components of the environment ecosystems according to the studied radionuclides.

The ^{90}Sr transfer factors in soils under irrigated crops decreased by 1.6 times for each studied object in the following row: soil under rice (99) > soil under alfalfa (61) > soil under corn (38) (Table 2, Figure 6). During the study, the accumulation of ^{90}Sr was observed in the irrigated lands of Crimea over time. The increase in ^{90}Sr concentration by 13% was noted in the soil under alfalfa at station 4A from 1994 to 1995 (Table 2, Figure 7).

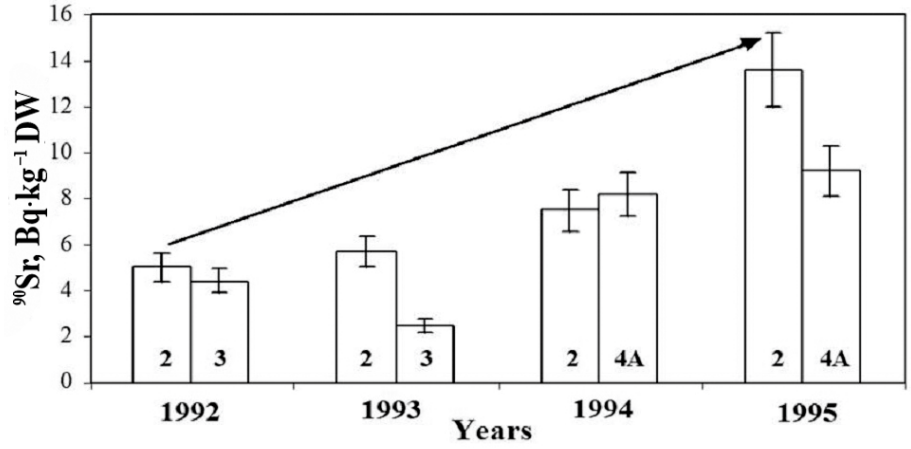

Figure 7. ^{90}Sr concentrations in soils under alfalfa (0–5 cm layer) in 1992–1995 at various sampling stations (stations 2 and 3—sampling station on the main canal, as 4A—sampling station on the outlet canal).

For the entire period of radioecological monitoring (1992–1995) in the soil under alfalfa at station 2, the concentration of ^{90}Sr in the 0–5 cm layer increased by 70%.

The investigations to study the transfer of post-accident ^{90}Sr to agricultural crops grown on irrigated lands along the NCC were carried out in 1992–1995 (Table 3, Figure 8). The research results showed that the accumulation of ^{90}Sr in alfalfa was on average 2–5 times higher than that for other crops (Table 3, Figure 8). The transfer factors of this radionuclide from irrigated soil to plants were calculated on the basis of the concentra-

tions of ^{90}Sr in soils and crops (Table 4). The highest value of TF in comparison with all the studied plants was also obtained for alfalfa (Table 4).

Table 3. Concentrations (C) of ^{90}Sr in cultivated plants collected from fields irrigated by the Dnieper water in 1992–1994.

Plant Characteristic	№ Station	Year	C,^{90}Sr, Bq·kg^{-1} ± σ, Dry Weight
Medicago sativa (alfalfa)			
Flowering plant		1992	6.3 ± 0.4
Shoots 3–4 cm	2	1993	1.5 ± 0.2
Flowering plant		1994	5.3 ± 0.3
	4A	1994	9.4 ± 0.6
Flowering plant	5	1994	7.7 ± 0.5
	5A	1994	8.1 ± 0.6
Triticum durum (wheat)			
Stems with spikelets	2	1992	1.5 ± 0.3
Zea mays (corn)			
Stems, leaves	2	1992	1.9 ± 0.2
	3	1992	0.8 ± 0.2
	4	1992	1.7 ± 0.3
		1994	1.9 ± 0.3
	5	1994	2.8 ± 0.3
	5A	1992	2.7 ± 0.3
Oriza sativa (rice)			
Whole plant	4	1992	5.5 ± 0.3
		1994	4.3 ± 0.3
Straw	5	1992	2.1 ± 0.3
Stems with grain		1993	0.7 ± 0.2
Straw		1994	2.2 ± 0.2
Stems with grain	4A	1993	1.5 ± 0.2
Straw		1993	1.4 ± 0.2
Straw	5A	1993	1.4 ± 0.2

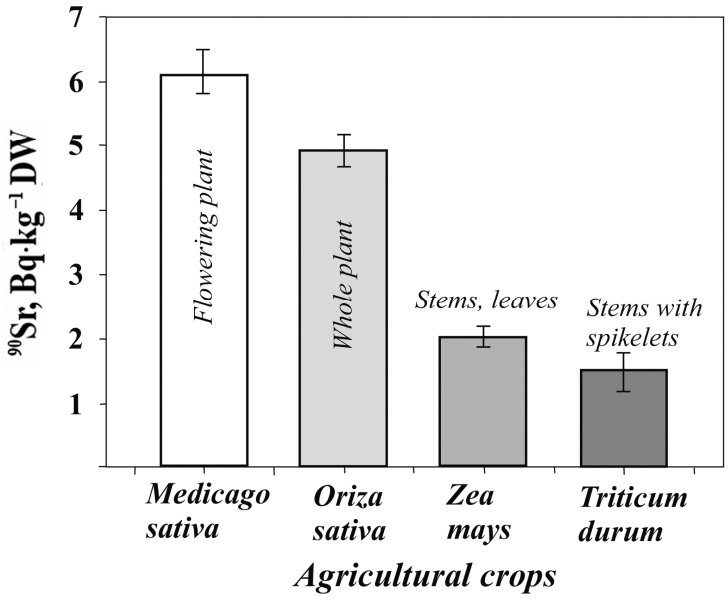

Figure 8. Average concentrations of ^{90}Sr in agricultural plants grown in irrigated fields along the NCC (1992–1995).

Table 4. Transfer factors (TF) of ^{90}Sr from irrigated soils to agricultural plants.

Station	Plant Name	TF, $m^2 \cdot kg^{-1}$
№ 2, 28 km of the NCC	alfalfa (green mass)	1.3×10^{-2}
	corn (stems, leaves)	3.7×10^{-3}
	wheat (straw)	3.0×10^{-3}
№ 3, 84 km of the NCC	corn (stems, leaves)	4.9×10^{-3}
№ 4, 125 km of the NCC	corn (stems, leaves)	4.3×10^{-3}
	rice (green mass)	7.5×10^{-3}
№ 5, 150 km of the NCC	corn (stems, leaves)	3.8×10^{-3}
	alfalfa (green mass)	2.1×10^{-2}

The determined TF for ^{90}Sr (Table 4) allow crops to be arranged in the following descending row: alfalfa (1.7×10^{-2}) >rice (7.5×10^{-3}) >corn (4.1×10^{-3}) >wheat (3.0×10^{-3}).

It was noted that the concentration of ^{90}Sr in the water of the NCC and cultivated agricultural plants was below the permissible levels used in the Russian Federation, regardless of the research period [36].

3.2. ^{137}Cs and Radionuclides of Transuranium Elements in the NCC Irrigation System

In 1990–1992, the activity concentrations of the "Chernobyl" radionuclides in the Dnieper water used for irrigated agriculture through the NCC had low values [14,37,38], which led to a much safer radiation situation in the region, in particular with respect to ^{137}Cs, compared with the situation on the territories located to the north and west of the ChNPP. It is known that the pre-accident level of the ^{137}Cs concentration in the Dnieper river water was about 3.7 $Bq \cdot m^{-3}$ [38].

In 1989–1995, the activity concentration of ^{137}Cs in the samples of the Dnieper water at station 1 remained practically at the same level (4.0–4.2 $Bq \cdot m^{-3}$), while in the water of the Dnieper reservoirs from the Kanevskoye to the Kakhovskoye in 1992, this value varied from 50 to 10 $Bq \cdot m^{-3}$ [37,39]. The concentrations of ^{137}Cs in the water of the irrigation and outlet canals were below the detection limit.

The results of the $^{239+240}$Pu activity concentration determination in suspended matter of water at different distances from the beginning of the NCC are shown in Figure 9.

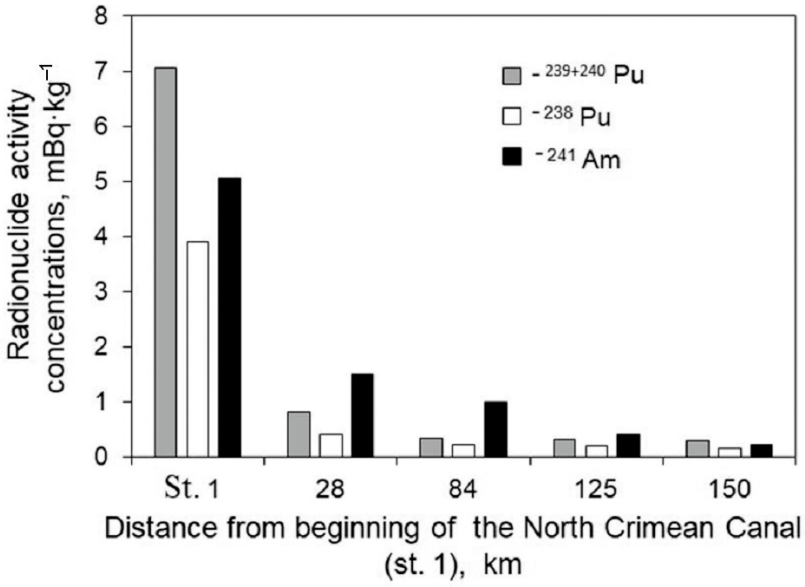

Figure 9. Changes in the activity concentrations of technogenic radionuclides in the suspended matter (d.w.) of the Dnieper water with increasing distance from the beginning of the NCC.

Their values decreased as the distance from the beginning of the NCC increased. About 95% of the plutonium was associated with suspended matter. In 1992, the activity concentrations of ^{238}Pu and $^{239+240}$Pu in the suspended matter of the Kakhovskoye reservoir water varied from 3.34 to 4.50 mBq·kg^{-1} and from 6.47 to 7.65 mBq·kg^{-1} of dry weight, respectively [17]. The decrease in the activity concentration of plutonium and americium radionuclides probably was observed with a distance from the Kakhovskoye reservoir since radionuclides are partially accumulated by bottom sediments. With active pumping of water, the bottom sediments of the canal were stirred up, and thus the suspension from the Dnieper River is diluted with less polluted sediment from the canal. The field irrigation regime could have influenced this parameter too. These processes were likely to lead to a decrease in the activity concentration of radionuclides of plutonium and americium in suspended matter in the canal with a distance from the Kakhovskoye reservoir.

No dependence of the plutonium isotope activity concentration in the soils of irrigated fields on the distance of the sampling stations (2, 3, 4) from the beginning of the canal was noted. Its value for $^{239+240}$Pu varied from 0.13 to 0.06 and to 0.17 Bq·kg^{-1} [19], which may have been due to the patchiness of radioactive fallout and the high sorption activity of soils and bottom sediments of the Dnieper reservoirs with respect to plutonium, and also, in general, a much lower level of activity concentrations of plutonium radioisotopes compared to cesium radioisotopes. The activity ratio of plutonium isotopes ^{238}Pu/$^{239+240}$Pu confirmed their Chernobyl origin, since it was about 0.5. Moreover, it is known that the composition of plutonium radioisotopes from the Chernobyl release was characterized by the ratio ^{238}Pu/$^{239+240}$Pu close to 0.4–0.6 [4]. For global fallouts of the northern hemisphere, this indicator when averaged was 0.036 [4]. Sampling of soils of a fixed area (0.05 m^2) at stations 2, 3, and 4 allowed for the estimation of the integral content of plutonium isotopes in the 0–5 cm layer. In this layer, the plutonium radioisotope inventories were 1.4–3.8 ^{238}Pu Bq·m^{-2} and 2.8–8.0 $^{239+240}$Pu Bq·m^{-2}.

Earlier studies of irrigated lands have shown that during irrigation of agricultural lands, part of ^{137}Cs was extracted by biotic and abiotic components of these ecosystems [40–42]. From the results presented in Figure 10, it follows that both types of agricultural irrigation, namely, sprinkling and full flooding of checks, which were used for rice cultivation, affect the values of the ^{137}Cs activity concentration in the soils on these fields.

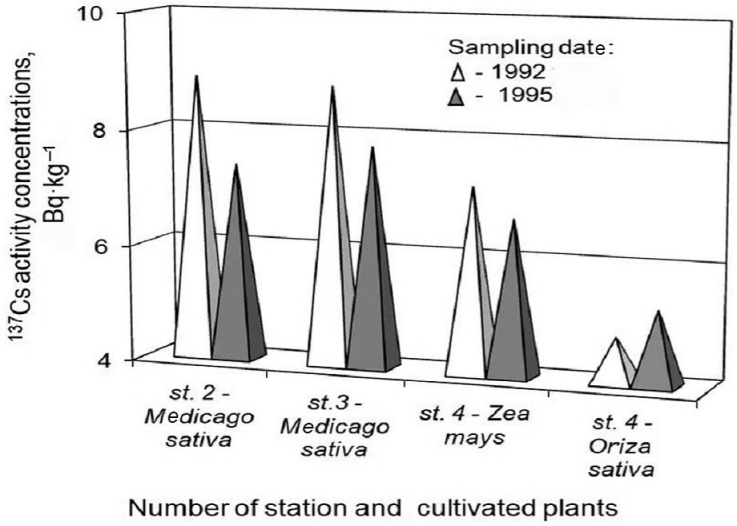

Figure 10. The ^{137}Cs activity concentration in irrigated soils (d.w.) under various crops.

The determination of radionuclides in the green mass of cultivated plants showed that the accumulation levels of ^{239,240}Pu were low and varied from 65 to 77 mBq·kg^{-1} (d.w.) in *Medicago sativa*, and from 0.2 to 2.5 mBq·kg^{-1} (d.w.) in *Oriza sativa*.

The vertical distribution of ^{137}Cs in the irrigated soil under *Medicago sativa* (Figure 11) testified to the deepening of the peak of this radionuclide in the soil. The distribution of ^{137}Cs in soils under *M. sativa*, which we selected in the same field at a distance of 50–100 m from each other, was similar in appearance: the maximum of ^{137}Cs was determined at a depth of 5–10 cm. The obtained soil profiles showed that the maximum values of the ^{137}Cs in irrigated soils were in the intermediate layers of the vertical soil profile. A similar distribution of radioactive cesium was noted in the soil under *M. sativa* in 1990 [14]. The vertical profile of ^{137}Cs in the virgin soil in the region of the NCC was also studied (Figure 11).

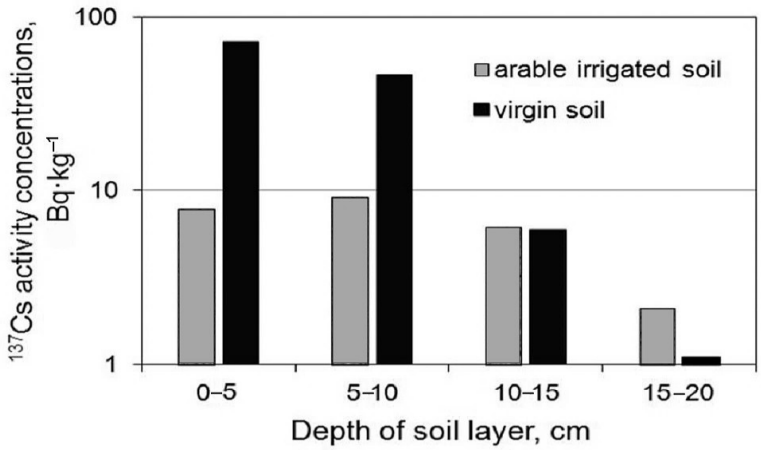

Figure 11. Vertical distribution of the ^{137}Cs activity concentration in the irrigated soil (d.w.) under *Medicago sativa* and in the virgin soil (d.w.).

Studies of the $^{239+240}$Pu activity concentration in alfalfa and corn at test sites 1 and 2 showed that the transfer factors (TF) of plutonium from soils to plants were equal (77–94) $\times$ 10^{-2} for alfalfa, and for corn, they were lower—(0.1–2.5) $\times$ 10^{-2} m^2·kg^{-1}.

The levels of activity concentration of cesium and plutonium radionuclides (as pedotropic elements) in bottom sediments of the NCC deserve no less attention. Bottom sediments that were taken in the main canal at station 1 were coarsely dispersed, silted sands, and at stations 2–5, they changed from silty sands to clayey silts. Bottom sediments in the irrigation canals consisted mainly of coarse particles with a small amount of admixture of plant remains. In the outlet canals, bottom sediments consisted of silts with a large amount of admixture of plant remains. The study of bottom sediments of the NCC showed that for them, there was a general trend in the spatial distribution of ^{137}Cs and plutonium isotopes—a decrease in their activity concentrations with distance as the sampling points moved away from the place where water entered the main canal of the NCC (Figure 12).

For an integral assessment of the ^{137}Cs and $^{239+240}$Pu removals from the Kakhovskoye reservoir to the Karkinitsky Bay of the Black Sea, the activity concentrations of these radionuclides in the bottom sediments of the NCC in this study were used. This reflected the general trends in their redistribution during the movement of the Dnieper water through the main canal and its branches. In this regard, a comparison was made of the ^{137}Cs and $^{239+240}$Pu activity concentration in the bottom sediments of the irrigation canals (through which water was taken to test sites 1 and 2), and in the outlet canals (through which water, after irrigation of the fields, was discharge into the Karkinitsky Bay of the Black Sea) (Figure 13).

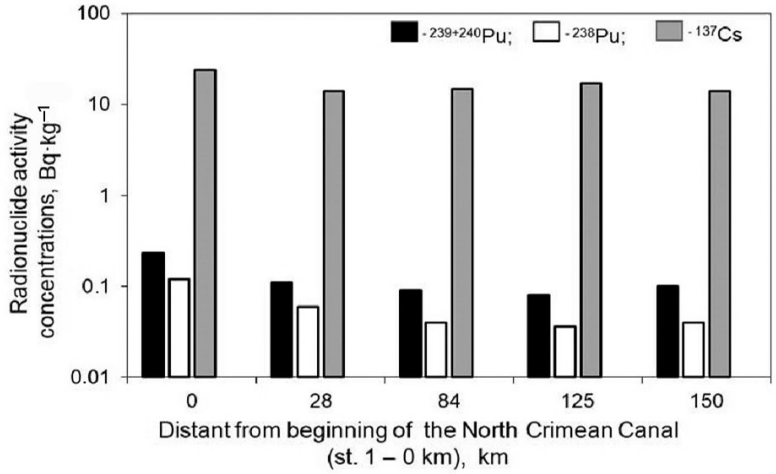

Figure 12. Change in the activity concentration of radionuclides in the bottom sediments (d.w.) of the NCC with increasing distance from the beginning of the NCC.

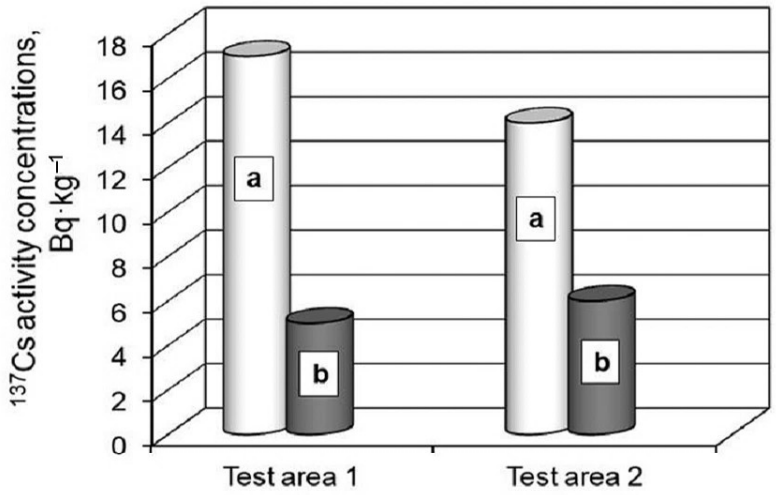

Figure 13. The ^{137}Cs activity concentrations in bottom sediments (d.w.) from the irrigation canal (a) and from the outlet canal (b) of test site 1 in the Krasnoperekopsky region and test site 2 in the Razdolnensky region of Crimea.

The activity concentration of radiocesium decreased in these canals at test site 1 from 5 to 17 Bq·kg^{-1} and at test site 2 from 6 to 14 Bq·kg^{-1}.

The studies of the ^{137}Cs concentration activity in water, bottom sediments of the NCC, the virgin soils adjacent to the canal, and the soils from irrigated fields to determine the contemporary background levels of technogenic radionuclides after the resumption of the Dnieper water supply to the NCC in the spring of 2022 were carried out.

Water samples taken in the main canal of the NCC in front of station 4 contained 1 Bq·m^{-3} of the ^{137}Cs activity concentration in dissolved form. In virgin soils adjacent to the canal, the activity concentration of ^{137}Cs in the 0–5 cm layer of soil was 13 ± 2 Bq·kg^{-1}; in the 5–10 cm layer, it was 15 ± 1.5 Bq·kg^{-1}; and in the NCC sediment, it was 21 ± 2 Bq·kg^{-1}. The $^{249+240}$Pu activity concentration in the sediments was 36.6 ± 5.9 mBq·kg^{-1}, and the ^{238}Pu activity concentration was below the detection limit. The activity concentrations of plutonium in irrigated soils (0–10 cm) from wheat field were 97.1 ± 9.0 mBq·kg^{-1} and for cesium were below the detection limit. Such levels of activity concentrations of technogenic radionuclides do not pose a danger now due to the use of the Dnieper water for irrigation, but in the future, observations on these values will be continued.

4. Discussion

4.1. ^{90}Sr in the NCC Ecosystem–Irrigated Soils–Cultivated Plants

The obtained results (Figure 2) showed that the Dnieper water, flowing through the NCC, is a radioecological factor of the long-term supply of the "Chernobyl" radionuclides from area of the ChNPP accident along the Dnieper River, through the cascade of the Dnieper reservoirs in the NCC, and along the canal, to the territory of Crimea and the irrigated agricultural lands of this region. The generalization and analysis of previously obtained results on the behavior of post-accident ^{90}Sr in the system "irrigated soils–cultivated agricultural plants" is important for planning future agricultural work in Crimea, especially with the resumption of rice cultivation in the region.

It was determined that the concentrations of ^{90}Sr in the outlet waters of the NCC (stations 4A and 5A, Figure 2) were 1.7–3.4 times less than in irrigation water supplied to irrigated agricultural lands (stations 4 and 5) (Figure 3). This was probably due to the withdrawal from the irrigation water and the deposition of an average of 51% ^{90}Sr in the irrigated soil and living components of the irrigated field, which indicated the ingress of ^{90}Sr from the water into the soil, as well the radioactive contamination of the irrigated fields of Crimea. At the same time, the concentration of ^{90}Sr in sea water in the Karkinitsky Bay of the Black Sea (station 5AA) was 3.8 times lower than in the outlet canal of the NCC (Figure 3). Those hydrological and biogeochemical self-purification processes contribute to a decrease of 3.8 times of the ^{90}Sr radioactive contamination in this area of the sea.

According to our calculations, 5.9 TBq of ^{90}Sr came with the Dnieper waters to the irrigated lands of the Kherson region and Crimea for the period of 1986–1999 (Table 1), which was about 7% of the total amount of this radionuclide carried from the Dnieper to the Black Sea [6].

The concentration of ^{90}Sr in irrigated soils under rice was on average only 23% lower than in bottom sediments, corresponding to the nearest NCC stations and the same sampling year. This testified to the same sorption properties of bottom sediments in the canals and soils of rice paddies and that the main source of the ^{90}Sr contamination of irrigated areas of the Crimean region is associated with the water migration of this radionuclide from the ChNPP accident area to significantly remote regions, such as Crimea.

During the study, the accumulation of ^{90}Sr was observed in the irrigated lands of Crimea over time. From 1994 to 1995, a 13% increase in ^{90}Sr concentration was noted in the soil under alfalfa at station 4A (Table 2). For the entire period of radioecological monitoring (1992–1995), the concentration of ^{90}Sr increased by 70% in the soil (0–5 cm layer) under alfalfa (station2) (Table 2).

It was determined that the accumulation of ^{90}Sr in cultivated plants did not depend on the remoteness of their cultivation sites from the beginning of the NCC. At the same time, interspecies differences were observed in the degree of accumulation of the ^{90}Sr by irrigated crops. The investigated cultivated plants can be arranged according to the degree of decrease in the concentration of ^{90}Sr in the following row: alfalfa > rice > corn = wheat (Table 3, Figure 8), which is in good agreement with the obtained results by other researchers. Thus, it was revealed [38] that ^{90}Sr is absorbed by legumes 2–6 times more intensively than by cereals. It was shown by a number of authors [40–42] that agricultural crops in terms of ^{90}Sr accumulation can be arranged in the following order: alfalfa > oats > barley > winter wheat > sugar beet > corn > cabbage.

The summarized results of the research allow us to fulfill the forecast on the conduct of irrigated agriculture in Crimea after the supply of the Dnieper water through the NCC resumed in 2022. The forecast is based on the following assumptions: the ^{90}Sr concentrations in the water of the NCC and irrigated agricultural plants did not exceed the maximum permissible concentrations (MPC) for the whole period of research (1986–1995); the concentration of this radionuclide in the Dnieper water, which began to flow into the NCC again from 2022, corresponds to the levels of ^{90}Sr for the first years after the ChNPP accident. Then, it is to be expected that the concentrations of the ^{90}Sr in crops (rice, corn, alfalfa, wheat) cultivated on the Crimean fields will also not exceed MPC adopted for the

quality and safe use of food raw materials [36] in the next 10 years (since 2022 to 2032). The obtained results of the vegetation experiments with rice showed [41,42] that the salinity of the soil and irrigation water contributes to a decrease in the content of ^{90}Sr and ^{137}Cs in plants. In the future, when growing such a crop as rice in Crimea, it should be taken into account that it is advisable to cultivate rice on saline soils and irrigate the plants with mineralized water with a part of it discharged into the collector network.

4.2. ^{137}Cs and Transuranium Elements in the NCC Irrigation System

The results obtained on the redistribution of cesium and plutonium radionuclides in the NCC system made it possible to trace the trends in the activity concentration change of the studied radioisotopes in the components of irrigated systems. They allowed for the evaluation of the buffer role of the western part of the NCC irrigation system (in the Krasnoperekopsky region—test site 1, and in the Razdolnensky region—test site 2) in the entry of these radionuclides into the Black Sea through the Karkinitsky Bay.

The $^{238,239+240}$Pu activity concentrations in the suspended matter of the NCC water (Figure 9) sharply (25 times) decreased as the sampling sites moved away from the beginning of the NCC main canal. Values of the ^{238}Pu concentrations in suspended matter changed from 3.9 mBq·kg^{-1}d.w. (station 1) to 0.2 mBq·kg^{-1}d.w. (station 5), and for $^{239+240}$Pu, it varied from 7.1 to 0.3 mBq·kg^{-1}, respectively [16,17].

A similar direction of changes in activity concentration was also observed for ^{241}Am, for which the values of its activity concentration varied from 5.1 mBq·kg^{-1} (station 1) to 0.2 mBq·kg^{-1} (station 4) (Figures 1 and 9). The observed dependences were probably due to the high sorption ability of plutonium and americium in relation to particles of suspended matter and the sedimentation of suspended matter to the bottom of the main canal of the NCC as far as water moved in it.

Taking into account the pedotropic type of biogeochemical behavior of cesium in freshwater ecosystems, its behavior in soils, the concentrators of these elements, is of greatest interest. The observed variability in the activity concentration of ^{137}Cs in soils under different cultivated plants (Figure 10) can also be caused by different local levels of atmospheric fallout in the early period after the ChNPP accident. Spotting in the distribution of radioactive contamination by ^{137}Cs of the soil surface under the same crop was noted during the study.

Thus, the ^{137}Cs activity concentration varied from 5.9 to 7.9 Bq·kg^{-1} of dry weight in the surface layer of soil taken at the same time under alfalfa in the same field. At that, soils under alfalfa contained higher activity concentrations of ^{137}Cs compared to soil from rice paddies (Figure 10). Moreover, the soil from rice paddies contained less ^{137}Cs than the soil under corn. Despite the possibilityof the effect of the radioactive fallout spotting on the levels of the activity concentration of ^{137}Cs in soils, it can be concluded that technology features of rice cultivation has the strongest effect on the amount of ^{137}Cs in irrigated soils. The influence of agriculture on the vertical distribution of ^{137}Cs in the soil was most noticeable when comparing the profiles of its distribution in irrigated and virgin soils (salt marshes) (Figure 11).

The study of the surface layer of virgin soil showed that the highest concentrations of ^{137}Cs were confined to the 0–2 cm layer (102 Bq·kg^{-1}), which was more than two times higher than that in the 2–5 cm layer. The specific activity of ^{137}Cs in virgin soil in the 15–20 cm layer was below the detection limit (0.1 Bq·kg^{-1}), while in irrigated soils, it was up to 25% of its value in the surface soil layer.

Consequently, ^{137}Cs was "locked" in the surface layer of 0–2 cm in virgin soils, and the maximum of this radionuclide concentration was located there. Conversely, in irrigated soils, ^{137}Cs migrates faster and deeper into the soil and its maximum was observed at a depth of 5–10 cm. Thus, arable irrigated agriculture, on the one hand, contributes to the leaching of ^{137}Cs from soils, and on the other hand, leads to an increase in its migration deep into the soils.

With the existing large heterogeneity of the initial atmospheric contamination with ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am of irrigated soils, taking into account their properties, as well as crop rotation, irrigation, and other conditions, it was rather difficult to fully establish the total contribution of the irrigation system to the extraction of these radionuclides from the Dnieper water for the entire period after the ChNPP accident. Therefore, in our opinion, the activity concentration of these radionuclides in the bottom sediments of the NCC can serve as an integral assessment of the levels of pollution of the aquatic ecosystem of the NCC and an assessment of its role in the migration and redistribution of the radionuclides from the Dnieper water to the Karkinitsky Bay of the Black Sea [4,28].

From the results of determining the ^{137}Cs in the bottom sediments of the NCC, presented in Figure 13, it follows that in the period after the Chernobyl NPP accident (from 1986 to 1995), no complete extraction of radiocesium from the Dnieper water was observed when it was used for irrigation. This is because a high water flow rate does not contribute to the formation of appropriate conditions for the maximum possible sorption of ^{137}Cs from the aquatic environment. Therefore, almost 99% sorption of ^{137}Cs should not have been expected at the point where the Dnieper water from the Kakhovskoye reservoir entered the NCC, as was observed under static experimental conditions [4]. Under natural conditions, the extraction of ^{137}Cs by bottom sediments of the NCC occurs gradually as the Dnieper water moves through the irrigation system. Thus, the distribution of ^{137}Cs in the bottom sediments of the NCC integrally reflects the entire process of transfer of this radionuclide with the Dnieper water both in the main canal and in the irrigation canals of this water system.

On the basis of this approach, an assessment was made of the ^{137}Cs export to the Karkinitsky Bay of the Black Sea to characterize the buffer role of the western part of the NCC irrigation system in the input of radionuclides into this sea area. For such assessment, radioecological materials were used, obtained for bottom sediments from the main canal of the NCC and at test sites 1 and 2 with a known irrigation area and fixed volumes of water supplied to them. During the period 1992–1995, the average ^{137}Cs activity concentration in bottom sediments in the main canal of the NCC at station1 was 33.5 Bq·kg^{-1}. The ^{137}Cs activity concentration in bottom sediments at the point of water supply to test site 1 (station 4) averaged 17.5 Bq·kg^{-1} by 1995, which corresponded to almost 52% of its value at the beginning of the NCC. This ratio of ^{137}Cs activity concentrations in bottom sediments at stations 1 and 4 indirectly reflected the same trends of change in the ^{137}Cs activity concentration in the water of the canal at these stations. With the volume of water that was supplied to test site 1 in 1992 equal to 34.5 million m^3, the total activity of ^{137}Cs introduced with the Dnieper water to this test site (1380 ha) was about 71.7 MBq during the irrigation period per year.

In total, the 47.6 MBq of ^{137}Cs remained in the ecosystem components of the irrigated agricultural lands of this test site after irrigation in 1992. At the same time, the 24.2 MBq ^{137}Cs was discharged into the Karkinitsky Bay of the Black Sea with drainage water, which amounted to 33.7% of this radionuclide activity that entered this area from the main canal.

On the basis of the results of studies and quantitative assessments, it can be concluded that from the 100% ^{137}Cs entered to the irrigated area, 66% of cesium remained in the irrigated field and 34% of cesium entered the outlet canal and further into the waters of the Karkinitsky Bay of the Black Sea.

A similar approach and calculation were carried out for test site 2 (6500 ha) with the volume of the Dnieper water supplied to it being equal to 162 million m^3. It has been established that about 70% of the ^{137}Cs activity that has arrived was extracted by the components of this ecosystem, and the remaining part (30%) was carried along with discharged drainage waters to the Karkinitsky Bay. In general, for the period from 1986 to 1995, about 674.4 GBq of radioactive cesium isotopes came from the Kakhovskoye reservoir into the main canal of the NCC, of which, 505.8 GBq went to the irrigated agricultural land of the NCC irrigation system.

When calculating the removal of plutonium radioisotopes into the Karkinitsky Bay of the Black Sea, their activity concentrations in water were used. As is known, the main depots of transuranic elements, including plutonium, are bottom sediments and soils. It is in these ecosystem components that the main reserves of plutonium isotopes that have fallen into the environment are contained. Both for water and for bottom sediments as well as for soils, a decrease in the density of radioactive contamination by plutonium with a distance from the site of the Chernobyl accident was characteristic, but at the same time, the patchiness of the distribution of contamination, due to the history of radioactive fallout, was also registered.

The amount of $^{239+240}$Pu radioisotopes in the Dnieper water that entered the main canal of the NCC during 1990–1995 remained practically unchanged and amounted to 105 MBq in general over this period. The total activity concentration of plutonium isotopes in the Dnieper water varied from 5.6 mBq·m^{-3} at the place of its supply from the Kakhovskoye reservoir to the NCC to 3.2 mBq·m^{-3} at station 4, i.e., at the entrance to test site 1. The concentration of plutonium in the drainage waters at station 4A after completion of the irrigation cycle of this test site was 1.3 mBq·m^{-3}. Consequently, the activity of these radionuclides, which arrived at this site in 1992, was about 110.4 kBq. The total plutonium activity in drainage water was 44.85 kBq.

As can be seen, 59.4% of the initial amount of plutonium in water that arrived at test site 1 in 1992 remained in its ecosystem, while 40.6% was transferred with discharged water to the Karkinitsky Bay. Our calculations based on the data of 1990–1995 showed that the removal of plutonium isotopes through this area varied from 40 to 41% of their initial amount in the water supplied for irrigation.

Similar calculations for test site 2 testify to the same role of the irrigated area in the extraction of plutonium from the Dnieper water. Up to 60% of the plutonium that fell on the irrigated fields with irrigation water remained in the components of the ecosystems of these test sites. At the same time, about 4.2–4.7 mBq of plutonium isotopes arrived per 1 m^2 of irrigated soils. In general, for the period from 1992 to 1995, about 105 MBq of plutonium entered the main canal of the NCC, of which, 77.7 MBq went to irrigated agricultural land.

It was determined that 627 kBq of $^{239+240}$Pu was input at test sites of the western part of the NCC irrigation system. Of this amount, 60% of the $^{239+240}$Pu was accumulated by the irrigated field (soil and plants), and 40% entered the outlet canals and then into the Karkinitsky Bay.

Consequently, the obtained percentages of extraction and deposition of radioisotopes (^{137}Cs and $^{239+240}$Pu) by the main canal of the NCC and two test areas quantitatively characterize the buffer role of the western part of the NCC irrigation system in relation to the influx of ^{137}Cs and $^{239+240}$Pu into the Karkinitsky Bay of the Black Sea with the Dnieper waters.

5. Conclusions

After 36 years since the ChNPP accident, the Dnieper water flowing through the NCC is still a radioecological factor that ensures the long-term flow of "Chernobyl" radionuclides from emergency areas close to the ChNPP along the Dnieper and its cascade of reservoirs through the NCC to the territory of Crimea, particularly its agricultural land.The concentration levels of ^{90}Sr in the Dnieper water of the NCC in 2022 correspond to those for 1986–1987. It was noted that regardless of the observation period, the concentration of ^{90}Sr in the water of the NCC was 11–175 times lower than the MPC [34] but 4–63 times higher than the pre-accident levels. The concentrations of the dissolved form of ^{137}Cs and $^{239+240}$Pu in the Dnieper water that entered the channel of the NCC in 2022 were at the level of the detection limit.

Analysis of the results of radioecological monitoring on the entry of technogenic radionuclides after the ChNPP accident with the Dnieper waters into the NCC made it possible to establish trends of these radionuclide content changes in the water and bottom sediments of the canal, in terms of the sampling station distances from the beginning of the

NCC. Between 1986 and 1995, the activity of radionuclides ^{90}Sr, cesium, plutonium, and americium in the Dnieper water decreased exponentially throughout the entire irrigation system of the NCC. This was due to the extraction of radionuclides by the components of this system, primarily soils of irrigated fields and bottom sediments of the NCC, as well as plants from irrigated fields.

Differences in the degree of accumulation of radionuclides by different agricultural crops were revealed. In legume plants (alfalfa), the accumulation of ^{90}Sr and $^{239+240}$Pu was more intense than in cereals. The transfer factor (TF) of ^{90}Sr and $^{239+240}$Pu in agricultural plants in the system: "irrigated soil–irrigated crops" were determined. The ^{90}Sr and $^{239+240}$Pu TF values for alfalfa were n $\times$ 10^{-2} and n $\times$ 10^{-1}, respectively. Transfer factors for wheat, corn, and rice for ^{90}Sr were n $\times$ 10^{-3}, and for $^{239+240}$Pu they were about n $\times$ 10^{-2}. The average ^{90}Sr distribution coefficients (K_d) for irrigated soils were 99 for rice, 38 for corn, and 52 units for alfalfa; for ^{240}Pu these values were n $\times$ 10^4.

The irrigation system of the NCC retains 43–59% ^{90}Sr, 59–60% $^{239+240}$Pu, and 66–70% ^{137}Cs of the activity concentration of radionuclides entering irrigation fields with the Dnieper waters in the NCC. Thus, the NCC irrigation system plays the role of a buffer against the pollution of the Karkinitsky Bay of the Black Sea with technogenic long-lived radionuclides of Chernobyl origin.

The studies carried out in 1990–1995 and the obtained trends in the development of the radioecological situation in the NCC irrigation system after the ChNPP accident, as well as the levels of the activity concentration of technogenic radionuclides determined in 2022 in bottom sediments, water, soil, and suspended matter of the NCC, allow for the assumption that in the absence of an increase in the input radionuclides of the Chernobyl origin with the Dnieper river waters to the NCC, the levels of activity concentration of radionuclides of ^{90}Sr, ^{137}Cs, and $^{239+240}$Pu in cultivated crops (rice, corn, alfalfa, wheat) will not exceed the maximum permissible concentration (MPC), being accepted for assessment quality and safe use of food raw materials in the coming years. According to state regulations on radiation safety in the Russian Federation for food products MPC ($^{238, 239}$Pu and ^{241}Am) = 0.01 kBq·kg^{-1}, MPC (^{137}Cs) = 1 kBq·kg^{-1}, and MPC (^{90}Sr) = 0.1 kBq·kg^{-1}.

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