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Research Article

# The Research of Artificial Radionuclides Speciation in Surface Water at the Semipalatinsk Test Site Territory

**Zh Ye Zhapasheva*and AK Aidarkhanova**

Branch "Institute of Radiation Safety and Ecology" NNC RK,Kurchatov 071100, Kazakhstan

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## 1. Abstract

This research examined the speciation of artificial radionuclides in the surface waters at the Semipalatinsk test site territory. The speciation of 90Sr, 137Cs and 239+240Pu in the most polluted water bodies of the "Degelen", "Experimental Field" and "Telkem" test sites are determined by cascade filtration. In the researched waters, the main speciation of 90Sr is the dissolved matter. For 137Cs it is characteristic to be in a dissolved matter, in the coarse suspension and thin colloids. The radionuclide 239+240Pu can be in various speciation forms depending on the object being researched with a predominance of the dissolved matter and coarse suspension. Results allow an estimation the potential for migration and bioavailability of artificial radionuclides to be assessed.

## 2. Introduction

The role of water in human life and economic activity is extremely great. For humans, water is a vital resource, satisfying its physiological and economic needs. The rapid development of science and technology in the XX century led to a sharp increase in pollution of water resources.

In radioecology and radiation protection, the assessment of impact on the environment and risks related to radioactive contamination is made by measuring the total concentration of radionuclides provided that radionuclides are uniformly distributed in the environment. Today, we know that if radioactive particles and colloids are present, the contamination will be uneven distributed and the average bulk concentrations may fail to be representative of the real distribution of the radionuclides[1].

Without knowing distribution regularities and radionuclide migration dynamics in land and aquatic ecosystems it is impossible to correctly evaluate and predict radiation situation, develop rehabilitation measures for areas already contaminated. However, the scientific and specialized literature has highly little information on physical and chemical forms of radionuclides in the natural environment. It is these forms that define mobility and bioavailability of radionuclides and, respectively, a potential impact of these on human and the environment [2].

Aquatic environment is the major pathway of contaminants including radionuclides transferred to long distances. Artificial radionuclides can be contained in significant quantities in surface water bodies of the Semipalatinsk test site (STS) and pose a potential hazard for ecosystems and man while travelling beyond testing areas. Duration of transuranic elements half-life should also be taken into account, which reaches dozens of thousands years that causes them to stay long in the biosphere.

This paper covers research findings of $^{90}$Sr, $^{137}$Cs and $^{239+240}$Pu speciation in STS surface water.

Surface water of STS territory are represented by the followingwater body types:

- reservoirs of man-made origin;
- water bodies of natural origin;
- watercourses.

Man-made reservoirs are located at the territory of "ExperimentalField", "Balapan", "Telkem" and "Sary-Uzen" testing sites. These, as arule, are craters formed as a result from aboveground or undergroundnuclear tests filled with water. Water bodies of natural origin – smallnatural lakes some of which dry up by the middle of summer. The majorSTS watercourses are streamflows at the "Degelen" test site and theShaganriver which flows along the border of "Balapan" site and goes beyond the STS border.

***Corresponding Author (s):** *Zh Ye Zhapasheva, Branch "Institute of Radiation Safety and Ecology" NNC RK,Kurchatov 071100,Kazakhstan, E-mail: Zhapasheva_zh@nnc.kz*

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Summarized data on speciation of radioactive elements in water allows to make an objective assessment to migration ability of radionuclides. Data can also be used when developing practical recommendations for evaluating radioecological status of STS lands and predicting the level of radionuclides content in STS ecosystem components and adjacent territories.

## 3. Materials and Research Methods

Radionuclide species are defined according to their physico-chemical properties such as nominal molecular mass, charge properties and valence, oxidation state, structure and morphology, density, degree of complexation.

Radionuclides in the environment are often present in relatively low concentrations, and the concentrations of individual radionuclides species are usually too low to be determined directly. Thus, fractionation techniques are most often needed prior to the homogenisation, radiochemical separation and measurements[1].

Cascade filtration through a membrane filter makes it possible to segregate particles in the range of micron to millimeter. Filtration (0.45 μm) and ultrafiltration systems are used for segregating suspended matter and colloid particles in water in the range of nanometer to micron [1].

In order to isolate various speciation of radionuclides, a cascade of 6 filters was chosen, which allows isolation of suspended, dissolved speciation as well as colloids and pseudocolloids of different size. In the course of fractionation, an aqueous solution was sequentially run through filters of different mesh size – from 8 μm to 0.003 μm. At the last filtration stages for determining colloid speciation ultrafiltration membranes were used. The major characteristics of these is the capacity of retaining molecules of a certain size, which is expressed as a nominal segregated molecular mass meaning that 90% of uncharged ball shaped molecules of the denoted molecular mass will be retained in the filter [3]. Thus, 0.003 and 0.007 μm membranes segregate molecules of 10 and 100 kDa molecular mass, respectively. This notation for the values of the molecular mass to be segregated is conventional for filters of mesh size close to the size of substance molecules[3, 4].

Figure presents a procedure for a sequential cascade filtration in determining radionuclides speciation in water, which allow extraction of suspended substances, pseudocolloids, colloids of various size as well as low-molecular compounds of humic and fulvic acids, other components (**Figure 1**).

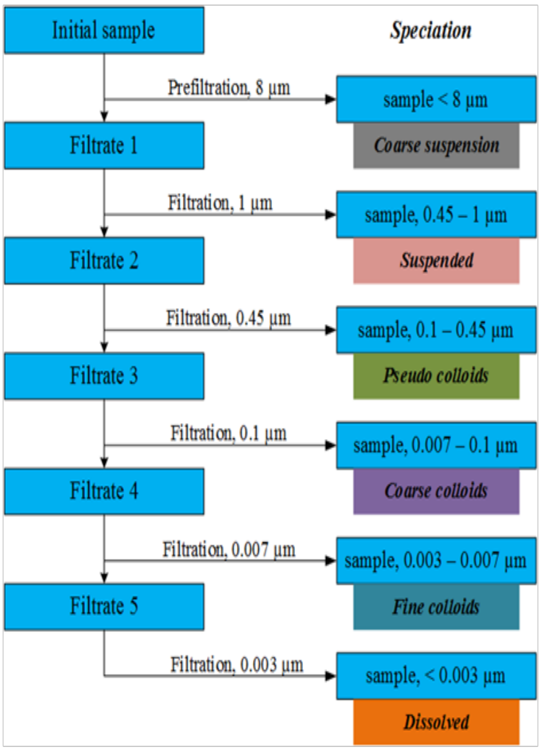


**Figure 1.**A sequential cascade filtration procedure for determining radionuclides speciation.

A choice of targets for research to study radionuclides speciation in water was determined based on results of earlier work [5, 6]. To research into mechanism of "colloid transfer", it was decided to study STS water bodies in more detail where maximum levels of $^{239+240}$Pu content in water was registered (higher than 0.1 Bq/l).

Water bodies of "Experimental Field" test site are craters formed asa result from aboveground and non-nuclear explosions filled with waterand in most cases overgrown across the perimeter with south cane(Phrágmitesaustrális). The diameter of the crater V-1 is 15 m.

"Telkem" site has 2 lakes formed as a result of excavation explosionsand filled with water. Telkem-2 was formed as a result of amultiple nuclear explosion 12.11.1968 г. Consisting of 3 charges inboreholes 2305, 2306 и 2307, and its shape is prolate currently havinga crater of about 120 m long and 60 m wide.

Within the"Degelen" site, several tunnels with either permanent or temporary brooks were found. These streamflows provoke takeout of radionuclides from the tunnels to the daily surface. The radionuclides transported out of the tunnels settle down in bottom sediments and form radioactive contamination along the streamflows [7 - 10].

Targets for research were streamflows of tunnels 104, 504 and 609 in "Degelen" site, a V-1 crater of the "Experimental Field" ground as well as water from the "Telkem-2" crater of "Telkem" site (**Figure 2**).

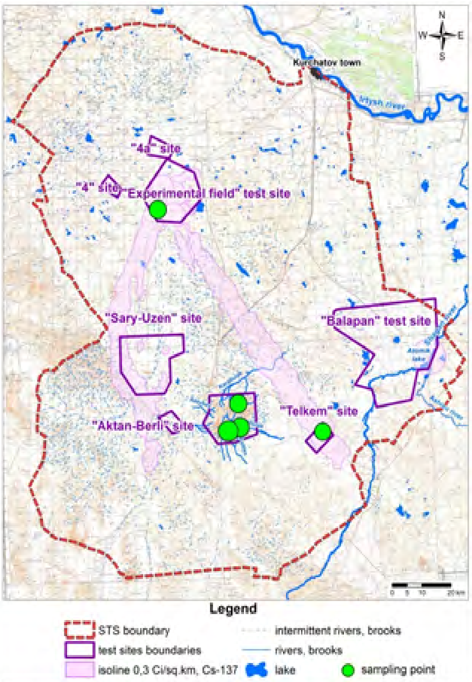


**Figure 2.**Schematic map of water sampling in the STS territory for research into determination of radionuclides speciation.

Water samples were collected according to(ST RK, 2003) avoiding spots of poor water exchange. The volume of water samples was 10 l. Samples were collected into clean polyethylene containers avoiding ingress of foreign impurities. Next, a water sample was filtered through a cascade of filters of the following mesh sizes - 1; 0.45; 0.1; 0.007 and 0.003 µm according to the procedure in figure (Figure 1).Experiments on cascade filtration were carried out directly on site, immediately after the sampling of water.

Once filtered, filtrate samples were collected and prepared through each cascade of filters for gamma- and beta-spectrometric measurements as well as 1 l aliquots were taken for preparing samples for the radiochemical analysis to determine $^{239+240}Pu$.

The content of anthropogenic radionuclides such as$^{137}Cs$,$^{90}Sr$,$^{239+240}Pu$ was determined in samples collected.

The water samples were concentrated with a coprecipitation technique: isotopes $^{137}Cs$ with copper hexacyanoferrate, $^{90}Sr$ with calcium carbonate and $^{239+240}Pu$ with ferrum hydroxide (III). The$^{137}Cs$ activity concentration was determined by γ-spectrometric measurements. The $^{239+240}Pu$ activity concentration was determined by the α-spectrometric analysis after extraction and chromatographic separation and electrolytic deposition. To account for the loss of plutonium during separation was injected $^{242}Pu$ as a tracer. A Canberra Co. manufactured α-spectrometer was used: it consisted of vacuum cells, α-radiation detectors, pulse analyzer and software GENIE 2000. The detection limits were calculated for each sample type and measurement time. The $^{90}Sr$ activity concentration was determined after radiochemical separation from the activity concentration of $^{90}Y$ by using the liquid scintillation beta-spectrometer "TRI CARB 3100 TR". To account for the loss of $^{90}Sr$ during analysis was used $^{85}Sr$ as a tracer. The yttrium chemical yield was determined by atomic emission spectrometry (ICP- AES)[11 – 15].

## 4. Results and Discussion

In **Table 1** are presented the physicochemical parameters and chemical composition of the studied water bodies of the STS.

As can be seen from the table, there are no significant differences in the degree of salinity between the studied water bodies. Except for the degree of mineralization of "Telkem-2" water which is classified as brackish. The water collected from the V-1 crater is fresh, as well as water from the streamflows of tunnels 104, 504 and 609. The pH values of water from streamflows of tunnels 104 and 609 are neutral, the water from the streamflows of tunnel 504 is slightly acidic and the water samples taken from "Telkem-2" and B-1 crater are slightly alkaline.

**Table 1.**The physicochemical parameters and chemical composition of the studied water bodies.

| Water bodies | pH | Mineralization, mg/dm³ | Total water hardness, mmol/ dm³ | Cation content, mg/dm³ | | | Anion content, mg/dm³ | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | $Na^{+}+K^{+}$ | $Ca^{2+}$ | $Mg^{2+}$ | $Cl^{-}$ | $HCO_3^{-}$ | $CO_3^{2-}$ | $SO_4^{2-}$ |
| V-1 crater | 8,0 | 690 | 5 | 180 | 50 | 40 | 240 | 270 | 12 | 90 |
| "Telkem-2" | 8,2 | 4320 | 41 | 1180 | 250 | 345 | 1430 | 180 | 9 | 2370 |
| tunnel 104 | 7,4 | 400 | 8 | 30 | 120 | 20 | 12 | 80 | 0 | 350 |
| tunnel 504 | 5,6 | 680 | 12 | 120 | 130 | 65 | 16 | 9 | 0 | 790 |
| tunnel 609 | 7,7 | 200 | 3 | 0 | 45 | 12 | 9 | 0 | 0 | 130 |

Speciation of artificial radionuclides $^{90}Sr$, $^{137}Cs$ and $^{239+240}Pu$ were considered separately for each object studied.Experimental research findings on speciation distribution of these radionuclides are presented as percentage of their total content in water researched[16].

All of the water bodies are found to have dissolved $^{90}Sr$ radionuclide, which indicates high migration capacity of this radionuclide.

Determinations of $^{137}Cs$ speciation in water of streamflows in tunnels 104, 504 and 609, as well as in water of the V-1 crater have shown that in water investigated has this radionuclides in the dissolved form figure (**Figure 4a**).

During the step-by-step filtration of "Telkem-2" water, $^{137}Cs$ activity concentration through each cascade of filters was at the level of 0.25±0.03 Bq/l, which corresponds to 89% of the initial content and is accounted for by the dissolved form. 7% of the total content of this radionuclide was in the coarse suspension, 4% in fine colloids figure (**Figure 4b**).

The analysis of findings showed that the major migration form of $^{239+240}Pu$ in water of tunnel 609 streamflow figure (Figure 5a) is a dissolved substance (60%). Coarse colloids and coarse suspension account for 16% and 12% of the total amount of activity. The content of $^{239+240}Pu$ as suspended substances in water of tunnel 609 streamflow was 6%, that of pseudocolloids – 4% and 2% for fine colloids.

Distribution of $^{239+240}Pu$ in water of "Telkem-2" showed that the predominant speciation of this radionuclides is a dissolved substance being 74% of the total activity, 19% is as suspended substance, 7% in the coarse suspension (**Figure 5b**).

Determinations of $^{239+240}Pu$ speciation in water of tunnel 504 streamflow have shown that 68% of this radionuclide is as a coarse suspension, 19% is in a suspended substance figure (**Figure 5c**). The remainder of the total content of $^{239+240}Pu$ in water of interest is distributed among colloids of different size and in the form of a dissolved substance (0.4%).

This distribution of $^{239+240}Pu$ by the speciations in the studied waters is affected by the ability of this element to hydrolyze at different pH values of water and to form soluble hydroxo complexes.

In water of the V-1 crater as well as in water of tunnel 104 streamflow the content of $^{239+240}Pu$ was below the detection limit of the technique used.

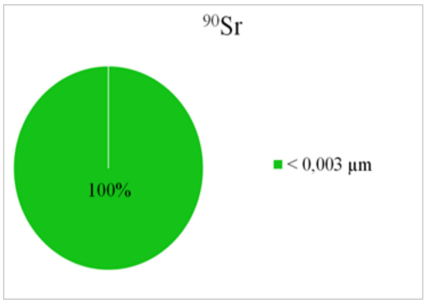


**Figure 3.** Distribution of $^{90}Sr$ radionuclide speciation in STS water bodies investigated.

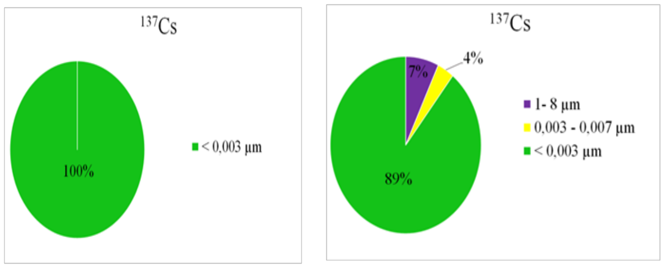


**Figure 4.** $^{137}Cs$ distribution by speciation in water of the V-1 crater and streamflows of tunnels 104, 504, 609 (a) and water of "Telkem-2" (b).

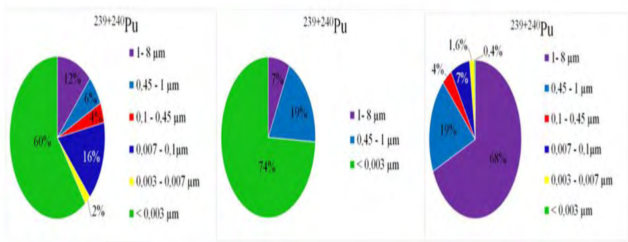


**Figure 5.** $^{239+240}Pu$ distribution by speciation in water of a streamflow in tunnel 609 (a), "Telkem-2" (b) and water of a streamflow in tunnel 504 (c).

## 5.Conclusions

Radionuclides distribution by speciation in STS surface water has been researched by a cascade filtration technique. Findings give an idea of migration capacity of artificial radionuclides in most contaminated water bodies of STS. High migration capacity is characteristic of $^{90}Sr$ which is found in a dissolved substance. The predominant speciation of $^{137}Cs$ in water investigated is a dissolved form, but there is also a distribution of this radionuclide in the form of coarse suspension and thin colloids. Speciation is inherent to $^{239+240}Pu$ in various forms dominated by dissolved and suspended ones. At that, the ratio of speciation and migration capacity depend on an object to be studied.

## References

1. Salbu B, Skipperud L. Speciation of radionuclides in the environment.Journal of Environmental Radioactivity. 2009; 281-282.

2. Novikov AP, Kalmykov SN, Tkachyov VV. Existing forms and migration of actinides in the environment. Russian Journal of Chemistry. 2005; 119-126.

3. Salbu B, Krekling T, Oughton DH. Characterisation of radioactive particles in the environment Analyst. 1998; 123: 843-849.

4. Ilyina SM. Role of organometallic complexes and colloids in the river runoff of the boreal climatic zone (the case of the Northern Karelia and Vladimir Meschera: a thesis by a cand. geol. and miner. sciences. – MGU. – Moscow. 2011; 186.

5. Kalmykov SN. Role of colloid particles in migration of actinides with ground water: author's abstract by dr. of Chemistry. Moscow. 2008.

6. Salbu B. Speciation of Radionuclides in the Environment Encyclopedia of Analytical Chemistry. 2006.

7. Lind OP. Transport of low 240Pu/239Pu atom ratio plutonium-species in the Ob and Yenisey Rivers to the Kara Sea Earth Planet. SciLett. 2006; 251: 33 - 43.

8. Buddemeier RW, Hunt JR. Transport of colloidal contaminants in groundwater: Radionuclide migration at the Nevada test site Appl. Geochemistry. 1988; 535 - 548.

9. Novikov AP. Migration and concentration of artificial radionuclides in environmental objects Geochemistry Int.2010; 48 (13): 1263-1387.

10. Aidarkhanova A K. Current contamination levels and nature of radionuclides distribution in the "water-bottom sediments" system in water bodies of the Semipalatinsk test site and adjacent territories / A.K. Aidarkhanova, S.N. Lukashenko Radiation Biology. Radioecology. 2017; 57: 286-296.

11. Aidarkhanova AK. Radionuclide transport in the "sediments – water – plants" system of the water bodies at the Semipalatinsk test site A.K. Aidarkhanova, S.N. Lukashenko, N.V. Larionova, V.V. PolevikJournal of Environmental Radioactivity. 2018; 184–185: 122-126.

12. Panistky AV, Lukashenko SN, MagashevaRYu. Characteristic features of radioactive contamination in natural environment components of tunnel streamflow ecosystems at Degelen massif Topical issues in radioecology of Kazakhstan. Pavlodar: Press House. 2010; 57-102.

13. Lukashenko SN. Topical Issues in Radioecology of Kazakhstan. Optimization of Survey of Semipalatinsk Test Site Territory for the Purpose of Transfer into Economic Turnover. Dom Pechati, Pavlodar. 2015; 87-93.

14. ST RK GOST R 51592-2003. "Water. General sampling requirements". Intr. 2005-01-01.

15. Measurement procedure №0307/3, 2001. A Procedure to Determine the Content of Artificial Radionuclides 239,240Pu, 90Sr and 137Cs in Natural Water byConcentration. Almaty, 7p (in Russian).

16. Measurement procedure with a gamma-spectrometer: MP 2143-91. Radionuclides activity in bulk samples. - Intr. 1998-06-02. - Reg. No. 5.06.001.98. – M. SPA VNIIFTRI. 1991.