

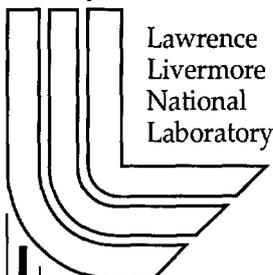
Analogue Study of Actinide Transport at Sites in Russia

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This article was submitted to
2003 International High-Level Radioactive Waste Management
Conference, Las Vegas, NV, March 30-April 3, 2003

February 12, 2003

U.S. Department of Energy



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Analogue Study of Actinide Transport at Sites in Russia

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ABSTRACT

The U. S. Department of Energy (DOE) and the Russian Academy of Sciences (RAS) are engaged in a three-year cooperative study to observe the behavior of actinides in the natural environment at selected disposal sites and/or contamination sites in Russia. The purpose is to develop experimental data and models for actinide speciation, mobilization and transport processes in support of geologic repository design, safety and performance analyses. Currently at the mid-point of the study, the accomplishments to date include: evaluation of existing data and data needs, site screening and selection, initial data acquisition, and development of preliminary conceptual models.

I. INTRODUCTION

Actinide speciation and transport data from Russian anthropogenic analogue sites can provide valuable corroboration of transport data, calculations and transport models used in the safety assessment of geologic repository concepts such as the Yucca Mountain Project (YMP), a proposed Russian repository, and possibly the repository programs of other nations. As an example related to the YMP, the concentration of mobilized actinides at the surface of exposed waste forms and the chemical speciation (i.e., specification of the oxidation state, chemical form, and physical state [surface precipitate, secondary mineral, or adsorbate]) during geologic transport are important uncertainties that are treated in a bounding fashion in current YMP analyses. Environmental analog data is one component, along with laboratory data and process modeling, of the approach to developing more accurate representations with narrower bounds. The results of this study may also contribute to international database development efforts and may support future uses of less conservatively bounded and more technically defensible models for radionuclide behavior.

II. OBJECTIVES

The major objective of this study is to obtain a better understanding of the interaction of actinides with the environment by using a number of contaminated sites and/or disposal facilities in Russia

as study locations and potential analogues for speciation, complexing, and migration or sequestration with emphasis on deep geologic environments. Transport parameters will be calculated, including diffusion coefficients in soil profiles, complexation strength of different organic and inorganic ligands with the actinides, chemical composition of soluble actinide species and colloidal fractions in natural waters (surface waters, underground waters and soil solutions). These parameters are important for both estimation of the safety of geological disposal and recovery of contaminated sites [1].

From the selected sites, data are being obtained on actinide speciation in natural surface and deep geologic ground waters, as well as in bottom sediments, soils, and in suspension, depending on the chemical and mineralogical composition of samples. These data form the basis for developing a mathematical model for prediction of the behavior of actinides in the environment. Data acquisition is being conducted in parallel with development of conceptual models for actinide behavior. The models developed for this project will be used in a confirmatory sense to test and corroborated process models used by the YMP to support performance assessments. It is clear that the Russian sites are not direct analogues for an engineered repository, nor for the Yucca Mountain site. However, the Russian sites can provide useful data on actinide behavior after decades of residence in the geosphere for comparison with results from process models used for repository evaluation.

III. SITE SCREENING

A number of sites are suitable for observation of environmental behavior of Np, Pu, and Am in Russia. Those considered in the site screening for this study included Mayak, Tomsk, and Krasnoyarsk.

At Mayak there is river, lake and basin sediment contamination from plant discharge, surface soil and water contamination from accidental atmospheric release, and subsurface migration from contaminated sediment in Lake Karachai.

Tomsk is a site where borehole injection of radioactive waste has taken place in isolated strata

At Krasnoyarsk, in addition to borehole injection of radioactive waste into isolated strata, there is river sediment contamination from plant discharge

Perhaps the best known site is located at Mayak [2] (near Kyshtym in Figure 1), where there is actinide contamination in soils, lake and river sediments, and surface and ground waters. High actinide activity ($10^2 - 10^7$ Bq/kg) in most components of the ecosystems allows valuable information to be obtained regarding speciation of actinides and their interaction with natural systems. A large amount of data exists on surface and shallow subsurface contaminants and limited information on deep subsurface migration.

Deep borehole injection of liquid high-, medium-, and low-level waste near Tomsk and Krasnoyarsk [2] (Figure 1) are also candidates for in-depth study. For more than thirty years, liquid radioactive wastes have been disposed of in geological formations at the Siberian Chemical Industrial Complex (SCC) at Tomsk and at the Krasnoyarsk Mining and Chemical Combine (MCC). Most of the injected waste consists of short-lived radionuclides, but some fission products with longer half-lives are also found. Although the actinide content in these wastes is low, these sites are of particular interest because of their large inventory (10^8 Ci), decades-long storage, and more than hundreds of meters of actinide migration in deep geologic strata.

A large volume of information has been collected over the decades on the chemical and radionuclide content of these sites. Less information is available on the speciation of actinides and mineralogical and physico-mechanical properties of ecosystem components. All of the sites are located in the temperate continental climate zone of the West Siberian Basin that receives 250–500 mm of precipitation annually, much of it as snow [2].

Each of the sites has certain advantages and limitations. For instance, Mayak has a very large number of control wells that are regularly sampled at different depth intervals. The source term is known. Relatively high concentrations of U, Np, Pu, Am, and Cm are found in groundwater samples that allow use of different speciation methods. Ground water compositions include a wide range of pH and Eh values. However, because of the shallow water table, temperature and pressure conditions of actinide migration may be more similar to those in saturated alluvium at Yucca Mountain than to conditions at depth.

The SCC at Tomsk has an advantage of extensive, ongoing data collection. The pre-injection conditions of radioactive waste are similar to what could be expected as background in natural waters at Yucca Mountain. Groundwater samples from control wells as far as 10 km away have high concentrations of anthropogenic radionuclides. Although the data are not currently available, SCC affords the possibility to collect samples from contaminated and uncontaminated locations within the same horizon. Disadvantages include preserving samples under the same thermodynamic conditions as are found at depth, and relative sampling cost.

Krasnoyarsk has advantages of extensive, ongoing data collection and multiple engineered and natural barriers. Disadvantages include the fact that only lower-level radioactive waste is injected into boreholes. Also even tritium concentration is not measurable in control wells in the low-permeability bedrock [3].

Based on the U.S interest in subsurface transport processes, data availability, and existence of other DOE programs evaluating certain of these sites, we selected the SCC injection site and subsurface contamination at PO Mayak for this study. Existing information for each site was summarized during the early phase of the study as a basis for site screening. Features and processes important to defining actinide speciation and transport, and initial conceptual models have also been evaluated.

Mayak

Mayak was the first enterprise launched by the former Soviet Union to reprocess fission materials (beginning in 1977) and to produce plutonium for military purposes (ending in 1987). Mayak is located between two rivers, the Techa and the Mishelyak. At present, two reactors remain in operation and five have been shut down.

During its 54 years of operation (since 1948), the activities at Mayak resulted in a local accumulation of large quantities of radiochemical wastes, with contamination of some parts of the region by long-lived radionuclides, mainly ^{90}Sr and ^{239}Pu [4]. Beginning in 1951, the main portion of nuclear waste was dumped into Lake Karachai, with a cascade of reservoirs and additional channels to reduce contamination of the Techa River. Lake Karachai (B-9 in Figure 2) is a natural closed water drainage basin that is connected to ground waters. The total radioactivity of the lake water is 2.0×10^9

Ci/l. The focus of our study at Mayak will be subsurface migration of actinides from these water reservoirs to different ground water horizons.

Tomsk

The SCC is located 20 km from Tomsk in a controlled area. Operation started in 1953 and it is now the largest plutonium, uranium, and transuranium element production complex in Russia. The complex includes plutonium production reactors, a radiochemical processing plant, an isotope separation plant and a chemical-metallurgical plant for nuclear materials. The industrial activities resulted in disposal of large quantities of liquid, aerosol, and solid nuclear waste.

Since 1967, radioactive solutions have been injected into the geological formations on a large scale. The total radioactivity of LLW injected into the deep geological repositories is 4×10^8 Ci. The chemical composition of the LLW is controlled by sodium salts, silica acid and radioactive isotopes of Sr, Cs, Ru, Ce, and others. The total radioactivity is 0.01 – 0.1 Ci/l in these wastes.

The injection horizons II and III (Figure 3) are situated above the low porosity horizon B and covered by impermeable horizon D. Horizons II and III are separated by clay lenses (C). The upper horizon IV serves as a buffer zone. Disposal horizons are low- and medium- grain size sandstones and clays [4]. The removal of radionuclides is achieved by passing through water-saturated sands (porosity about 32%). Effective porosity is estimated to be 12 % by volume. The "free" water is a calcium-sodium hydrocarbonate in composition. Water in the dead-end pores in the sand occupies about 20 % of the volume. This water is more basic in chemical composition than water in the connected pores.

The injected waste solutions are multicomponent systems containing acids or bases, salts, and complex forming substances. Radionuclides may be present as single ions, complexes, and as colloids or pseudocolloids. The huge difference in composition of the solutions and natural waters leads to different interactions in the rock-water system and a large degree of chemical heterogeneity.

IV. PRELIMINARY CONCEPTUALIZATION

A conceptual model of actinide behavior in natural environments should include the main processes that govern actinide migration, including

description of the geologic setting, radionuclide source, and transport processes (see Table 1). As this model develops, existing data is evaluated and further data needs are defined to guide field studies.

Mayak

Radionuclide transport in the deeper subsurface region at the Mayak site is primarily seepage of contaminants from the storage basins through interconnected fractures. The underground water system consists of a single aquifer 55 – 100 m thick; the bedrock contains fracture zones and sedimentary horizons. Continuous migration of contaminants into the saturated zone occurs through the bottom of B-9 (Lake Karachai) and B-17 (Old Swamp) where the contaminant plume displaces fresh waters. Because earlier studies [8] indicated negligible diffusion of transuranic elements (TUE) into minerals of the surrounding weathered porphyritic bedrock, the kinetics and thermodynamics of surface sorption will dominate TUE migration.

The connected reservoirs form a complicated multicomponent system with high salt content and biological activity. Their maximum depth is 8 m. The plutonium and americium content is low in waters of reservoirs B-10 and B-11 but is concentrated in bottom sediments to 100 Bq/g. The radioactivity of ^{60}Co , ^{90}Sr , ^{106}Ru , and ^{137}Cs in the saturated zone is 20 – 300 times lower than in waters of reservoir B-9; NO_3 content in a lens under the reservoir is 3.5 – 5 times higher than its concentration in water of the reservoir. Ground waters from different drillholes have a maximum in dissolved organic carbon of 0.5 mg/l and pH of 7.9 – 8.2. As a rule the concentration of radionuclides is from greatest to least: ^{90}Sr > ^{60}Co > ^{137}Cs > ^{239}Pu > ^{241}Am > ^{237}Np . In core samples, the same radionuclides were found as in the underground waters. As a rule, radioactivity decreases in core samples in the same manner as in the ground waters [4].

Radionuclide retardation during subsurface migration can be accomplished by sorption onto host rock or fracture lining materials through ion exchange or adsorption or by filtration of particulates.

Tomsk

The disposal of LLW by injection into deep boreholes within permeable horizons surrounded by low-permeability clay horizons began in Russia in the mid-twentieth century. The waste solutions are acidic or basic and differ significantly from groundwater compositions having nearly neutral pH and low

dissolved solid contents. The heat-generating radionuclides in the waste lead to higher temperatures (varying at the sites from 40–170°C) that increase the waste-rock-water interactions in a non-equilibrium system in which interactions between waste and host rock decrease with distance from the injection wells. The unique features of these injection sites are the high acidity or basicity of waste solutions, high sodium and nitrate content and high acetate and transition metal concentration in some solutions [3]. The fission products and actinide elements are present at microscale concentrations.

Rock in the exploration horizon is composed primarily of quartz, feldspar (10–20%), mica (2–10%), montmorillonite and kaolinite clays (up to 15%). The clays have high ion-exchange capacity. The sorption of radionuclides on the rock leads to temperature increases and transformation of both waste components and the surrounding rock. Removal of transition metal ions as oxides or hydroxides takes place by thermohydrolysis. These new phases have high sorptive capacity toward actinides, particularly plutonium. Radiolysis of acetates results in an increase of pH, decreasing the solubility of americium and neptunium.

The main "metasomatic" process that occurs at the site is argillization, which occurs at temperatures of 50–250°C. The increase in clay content leads to the transport of microcomponents of LLW to the solid phase.

Radionuclide removal from waste solutions can be accomplished by the following:

- sorption into rock through ion exchange or onto rock by adsorption
- co-precipitation with major cations of waste solutions
- sorption onto newly formed phases
- filtration of particulates

Different processes dominate, depending upon distance from the injection well. The maximum temperature is observed near injection wells due to chemical reactions between waste and host rock. The solutions migrate as wastes are injected (the rate of groundwater flow is about 3–5 m/yr). Because of the temperature gradient, the precipitation of low-solubility complexes occurs as products of mineral alteration. Regions further from the injection well are characterized by co-precipitation of radionuclides and the host rock is much less altered. At the border of the spreading waste plume, an intermediate mixing zone of waste solutions with groundwater is formed. The salt background decreases significantly in this zone

and radionuclides are sorbed at the non-disturbed rock surfaces.

Colloids could be formed upon neutralization of nuclear wastes in collector strata, sorption of radionuclides as colloidal silica, clay, microbes, humic colloids, etc. Possibly the colloids are unstable in high temperature solutions due to aggregation on solid phases. A temperature increase leads to decreasing colloid stability.

V. RESULTS TO DATE

As the first step in the field studies, water samples were collected and analyzed from boreholes at Mayak and the SCC-Tomsk.

Mayak

During the first sampling campaign, waters from the Lake Karachai zone were chosen. This zone was chosen because the composition of waters at the source of contamination is known, a variety of field and laboratory hydrological and geophysical investigations had been performed, and the main factors that govern the scale of radionuclide spreading are known.

Ground water and rock core samples were collected south of Karachai (Figure 2) in order to minimize the influence of reservoirs B-17 and B-4 that differ in radiochemical composition from Lake Karachai. Average ground water flow is estimated to be 0.2 m/day from 20-year observations and 0.17 m/day for B-9 reservoir solutions [6]. The migration of ⁹⁰Sr and ⁶⁰Co is estimated at 0.23 and 0.14 m/day, respectively. Because it was known that the maximal penetration of radionuclides is observed at the depth of 100 m [7], samples were collected from 90-100 m depth.

The data on radionuclide concentration were obtained without pre-concentration. All of the samples were micro- and ultrafiltered. It was noted that 60 to 80% of uranium was found in solution after ultrafiltration, in contrast to samples collected at the SCC (Table 2). pH ranged from 7.5 to 8.2, with an average of 7.9 for the 11 wells. Eh was 80–120 mV, (2 exceptions were 150 and 300), with an average of 100 mV.

For radionuclides in solution, the nitrate ion concentration provides a useful tracer for migration. Uranium concentration correlates reasonably well with nitrate concentrations (Figure 4), as most uranium is dissolved rather than particulate.

Colloidal transport is more important for the TUEs, for which nitrate concentration is a less reliable tracer.

SCC

At the SCC, three wells were sampled at different distances from the injection sites. Two of the wells are located inside the contamination zone (S-66, and A-44) and one (S-15) is outside this zone. The samples were collected from disposal horizon II (Figure 3). The composition of ground water measured at the S-15 well is shown in Table 3 along with the contrasting composition of oxidizing waters from well J-13 and UE-25p#1 at Yucca Mountain. The S-15 water predominantly a Na-bicarbonate, is slightly acidic and reducing, and has a high dissolved salt content. A radiochemical analysis was performed on samples from well S-15. The concentration of anthropogenic radionuclides (^{137}Cs , ^{241}Am , ^{237}Np , ^{60}Co , ^{154}Eu , and ^{90}Sr) were all below detection limits. Concentrations (in Bq/l) were determined for ^{40}K (90.15), ^{238}U (0.022), ^{226}Ra (0.0083), ^{228}Ra (0.014), and ^{228}Th (0.0060).

The ground water from S-66 and A-44 wells is contaminated by the spreading injection plume. The nitrate content is as high as 1 g/l. The concentration of cations is more than an order of magnitude higher than that for the S-15 well. The composition of the samples from S-66 and A-44 before and after filtration is shown in Table 4.

VI. SUMMARY

Sites in Russia for the study of actinide transport processes have been screened, and two sites selected for further study. Model development and data acquisition are being pursued in parallel with iteration between available data and model information needs. Existing information for each site has been summarized during the early phase of the study as a basis for site screening. Features and processes important to defining actinide speciation and transport, and initial conceptual models have also been evaluated during the site screening. Initial acquisition of new experimental data has begun and will continue to provide the information needed to fully develop the actinide behavior model.

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Acknowledgments

This work was performed under the auspices of the U.S. Department of Energy by University of California: Lawrence Berkeley National Laboratory, and Lawrence Livermore National Laboratory, under contract No. W-7405-Eng-48, and Vernadsky Institute of Geochemistry & Analytical Chemistry, Russian Academy of Sciences, via CRDF Project RG0-20102-RW40.

The authors thank the ANS reviewers for their efforts.

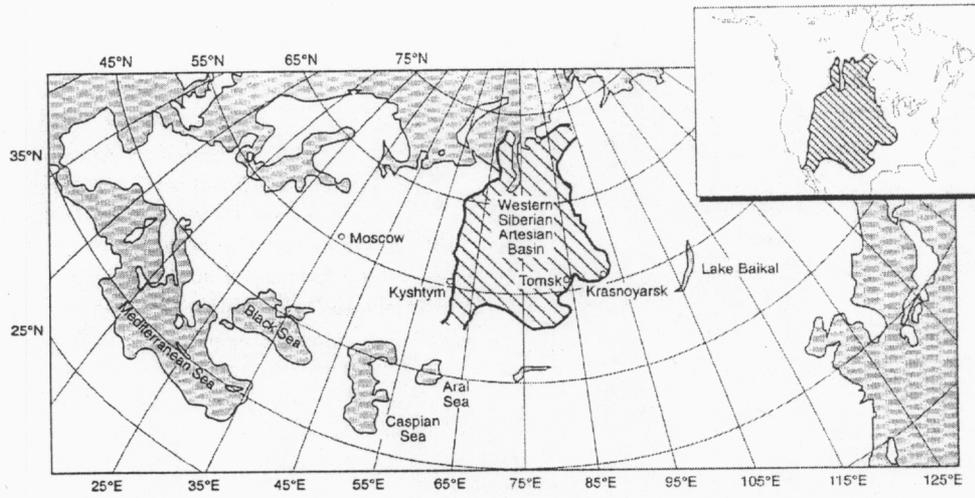


Figure 1. Index Map Showing the location of the West Siberian Artesian Basin and its size relative to North America (inset) [2]

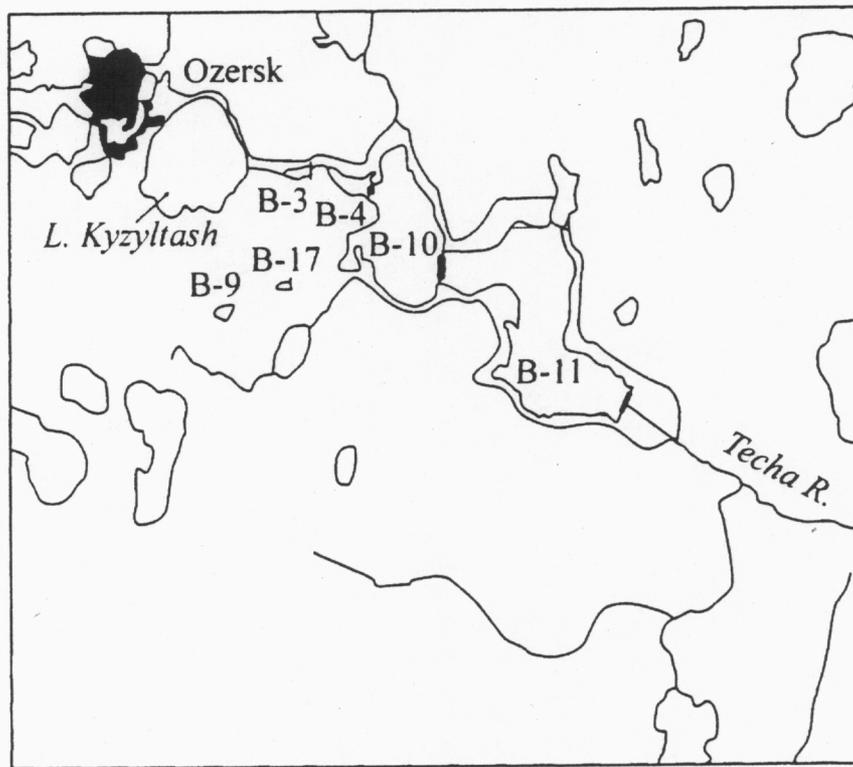
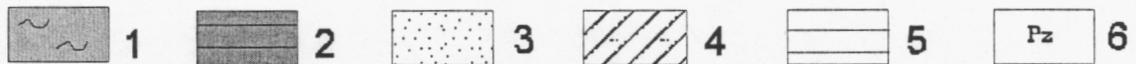
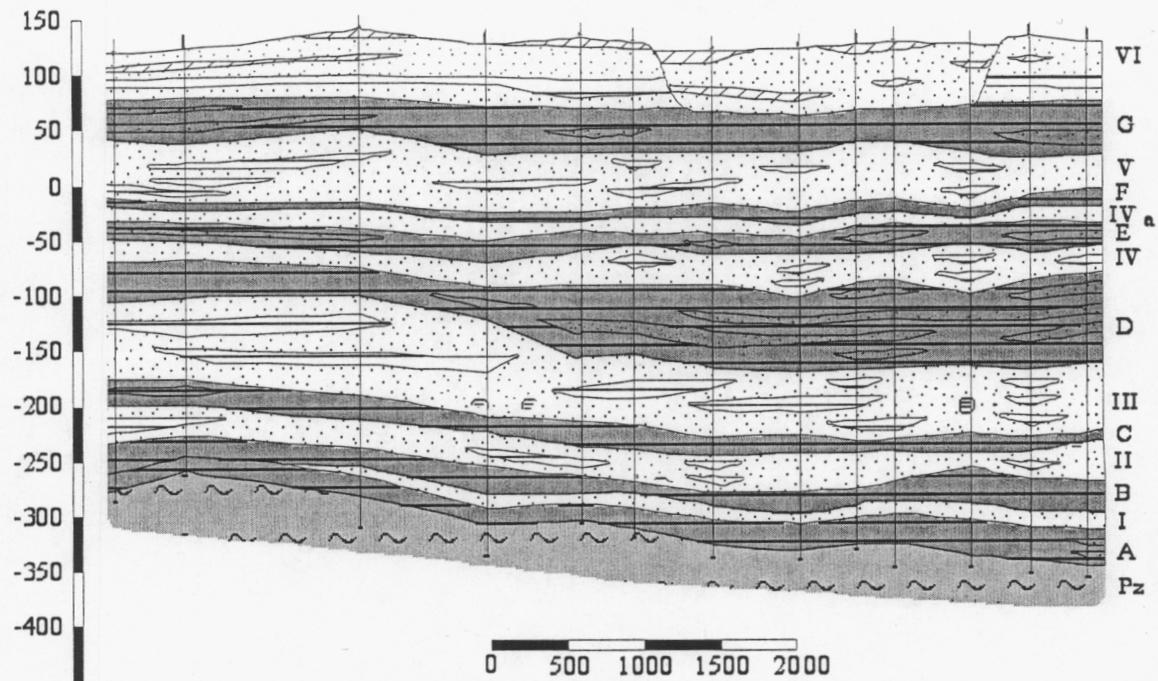


Figure 2. The cascade of industrial basins at the Mayak site [8]



Scale in meters

Figure 3. The geological profile of LLW in the underground burial site at SCC [4]

Note: Roman numerals indicate permeable horizons and Latin letters indicate low-permeability horizons. Symbols: 1 – Paleozoic sand and shale; 2 – low-permeability horizons; 3 – sand; 4 – loam; 5 – clay lenses; 6 – Paleozoic bedrock

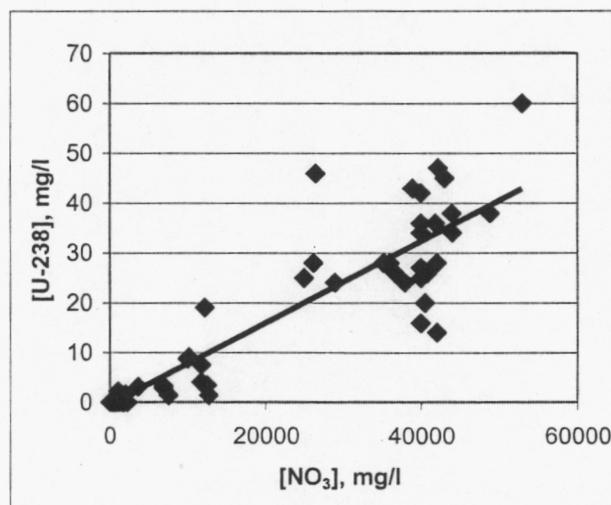


Figure 4. Nitrate concentration correlation with uranium in ground waters near Lake Karachai [5]

Table 1. Radionuclides and Features of Selected Study Sites

Site	Features	Radionuclides
Mayak	<ul style="list-style-type: none"> Vertical seepage from storage basin sediments Fracture network flow and transport Retardation and surface reaction processes 	Fission products: ⁹⁰ Sr, ⁶⁰ Co, ¹³⁷ Cs, ¹⁰⁶ Ru, Actinides: ²³⁸ U, ²³⁹ Pu, ²⁴¹ Am, ²³⁷ Np
Tomsk	<ul style="list-style-type: none"> Lateral transport from high pressure injection wells in permeable strata. Strong chemical interaction of waste and rock Dissolution and precipitation processes Radiation and decay heat effects 	Fission products: ⁹⁰ Sr, ¹³⁷ Cs, ¹⁰⁶ Ru, ⁹⁵ Zr, ⁹⁵ Nb, ¹⁴⁴ Ce Actinides: ²³⁸ U, ²³⁹ Pu, ²⁴¹ Am, ²³⁷ Np

Table 2. Results of Analyses of Water Samples from Wells near Lake Karachai [5]

Sample	Borehole #	pH	Eh (mV)	NO ₃ ⁻ (mg/l)	U (mg/l)	Sr-90 (Bq/l)	Cs-137 (Bq/l)	²³⁷ Np* (Bq/l)	^{239,240} Pu, Bq/l	²⁴¹ Am (Bq/l)	²⁴⁴ Cm (Bq/l)
1	63	-	-	35 200	28.0	97000	190000	9.1	1.59	6.40	14.0
2	65	7.5	300	10 100	9.1	7100	15.0	5.7	1.65	0.54	-
3	43	7.8	150	6 900	3.2	6650	<0.3	2.4	0.005	0.003	-
4	28	7.9	100	970	0.11	1230	<0.3	0.51	0.089	0.15	0.035
5	176	7.8	90	20	0.010	546	14.0	0.22	0.25	0.046	0.010
6	8	7.9	100	1.2	0.008	19.0	<0.3	0.04	0.35	0.032	0.008
7	20	8.2	80	2.0	0.012	862	3.5	0.02	0.061	0.081	0.012
8	1	7.9	100	≥1	0.012	936	3.8	-	0.15	0.010	0.010
9	13	7.9	100	≥1	0.012	66.0	<0.3	-	0.048	0.022	0.012
10	15	7.8	120	≥1	0.013	86.0	<0.3	-	0.17	0.16	0.035
11	14	7.9	100	≥1	0.006	236	<0.3	-	0.029	-	-

* - luminescent determination without concentration

Table 3. Comparison of Average Composition of Groundwater from S-15 Well with Waters from Two Yucca Mountain Wells [5]

Parameters	Dimension	Composition		
		S-15	J-13	UE-25p#1
pH		6.69	6.9	6.7
Eh	MV	-95	100	360
Salt content	mg/l	380	-	-
K ⁺	mg/l	5.	5.3	13.4
Na ⁺	mg/l	38.	45	171
NH ₄ ⁺	mg/l	0.08	-	-
Ca ²⁺	mg/l	19	11.5	87.8
Mg ²⁺	mg/l	12	1.6	31.9
Mn (total)	mg/l	0.07	-	-
Fe (total)	mg/l	3.2	0.4	<0.1
SiO ₂ (total)	mg/l	7.3	66	66
Al	mg/l	0.5	-	-
U	mg/l	1.4E-3	-	-
Th	mg/l	9.1E-5	-	-
HCO ₃ ⁻	mg/l	285	-	-
NO ₃ ⁻	mg/l	0.2	10.1	<0.1
NO ₂ ⁻	mg/l	0.05	-	-
Cl ⁻	mg/l	10.5	6.4	37
F ⁻	mg/l	0.12	2.1	3.5
SO ₄ ²⁻ (total)	mg/l	2.0	18.1	129
CO ₃ ²⁻ (total)	mg/l	289	118-143	960
β-activity	Bq/l	0.1	-	-
α-activity	Bq/l	0.05	-	-

Table 4. The Chemical Composition of S-66 and A-44 Well Water [4]

Element	S-66			A-44		
	Concentration in filtrate (mg/l) for six meshes					
	450 nm	50 nm	5 nm	450 nm	50 nm	5 nm
Li	0.018	0.018	0.016	0.017	0.017	0.017
Na	566.1	565.6	564.7	366.4	359.7	353.4
K	66.4	66.2	66.1	62.6	60.9	59.0
Cs	2.6E-4	2.0E-4	2.1E-4	1.9E-4	1.7E-4	1.5E-4
Ca	263.3	260.6	258.2	296.5	287.4	265.8
Mg	272.3	269.4	264.9	235.1	232.6	210.2
Ba	0.18	0.18	0.10	0.13	0.11	0.11
Sr	12.0	12.0	8.1	10.0	10.0	10.0
Al	0.36	0.35	0.19	0.28	0.19	0.058
Fe	0.40	0.12	0.07	0.096	0.033	0.016
Si	5.5	5.1	4.9	5.8	5.6	5.2
Mn	0.37	0.37	0.18	0.40	0.33	0.26
Eu	0.28	0.077	0.029	0.034	0.029	8.8E-3
Zr	3.1E-3	3.1E-3	8.0E-4	1.1E-3	0.74E-3	0.17E-3
Th	3.5E-5	3.4E-5	1.0E-5	6.2E-5	5.5E-5	3.1E-5
U	3.31	3.31	0.45	0.043	0.013	1.0E-5