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*«Қазақстан Республикасы Ұлттық ғылым академиясының Хабарлары. Геология және техникалық ғылымдар сериясы» ғылыми журналы 2016 жылдан бастап халықаралық реферативтік және ғылымиметриялық Scopus дерекқорында индекстеледі және тұрақты библиометриялық көрсеткіштерді көрсетіп келеді.*

*Сонымен қатар журнал Web of Science платформасының (Clarivate Analytics, 2018) халықаралық реферативтік және наукометриялық дерекқоры Emerging Sources Citation Index (ESCI) тізіміне енгізілген.*

*ESCI дерекқорында индекстелуі журналдың халықаралық ғылыми рецензиялау талаптары мен редакциялық этика стандарттарына сәйкестігін растайды, сондай-ақ Clarivate Analytics компаниясы тарапынан басылмды Science Citation Index Expanded (SCIE), Social Sciences Citation Index (SSCI) және Arts & Humanities Citation Index (AHCI) дерекқорларына енгізу қарастырылуда.*

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*Научный журнал «News of the National Academy of Sciences of the Republic of Kazakhstan, Series of Geology and Technical Sciences» с 2016 года индексируется в международной реферативной и наукометрической базе данных Scopus и демонстрирует стабильные библиометрические показатели.*

*Журнал также включён в международную реферативную и наукометрическую базу данных Emerging Sources Citation Index (ESCI) платформы Web of Science (Clarivate Analytics, 2018).*

*Индексирование в ESCI подтверждает соответствие журнала международным стандартам научного рецензирования и редакционной этики, а также рассматривается компанией Clarivate Analytics в рамках дальнейшего включения издания в Science Citation Index Expanded (SCIE), Social Sciences Citation Index (SSCI) и Arts & Humanities Citation Index (AHCI).*

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## ASSESSMENT OF THE RADIOCAPACITY OF ATOMIC LAKE AT THE SEMIPALATINSK NUCLEAR TEST SITE

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**Abstract.** *Relevance.* The Atomic Lake, formed by an underground thermonuclear explosion at the Semipalatinsk Nuclear Test Site in 1965, remains a radioactively contaminated water body. Radionuclides deposited in the surrounding catchment area can be redistributed and transported into the lake by surface runoff, requiring assessment of their long-term dynamics. *Objective.* The study aimed to assess changes in the radiocapacity of the inner and outer Atomic Lakes by analyzing the dynamics of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  in their catchment areas between 2014 and 2020, considering radioactive decay. *Methods.* Monitoring data from 2014 and 2020 obtained by field and laboratory gamma spectrometry were analyzed. Catchment boundaries were delineated using QGIS. Total radionuclide activities

were calculated from specific activity values, the number of contaminated sectors, and soil-density parameters. Temporal changes were evaluated considering the half-lives of the radionuclides. *Results.* The calculated catchment areas were 0.16 km<sup>2</sup> for the inner lake and 3.07 km<sup>2</sup> for the outer lake. From 2014 to 2020, the calculated total activity of <sup>137</sup>Cs decreased from 2.169×10<sup>9</sup> to 1.158×10<sup>9</sup> Bq, <sup>241</sup>Am from 2.906×10<sup>8</sup> to 2.663×10<sup>8</sup> Bq, and <sup>152</sup>Eu from 1.018×10<sup>9</sup> to 5.288×10<sup>8</sup> Bq. After accounting for radioactive decay, the calculations indicated redistribution and loss of radionuclides from the investigated catchment areas, with differences among radionuclides and between the inner and outer lake systems. *Conclusions.* The results demonstrate changes in the radionuclide inventory of the Atomic Lake catchment system between 2014 and 2020 that cannot be explained by radioactive decay alone. The proposed approach can be used to assess long-term radionuclide redistribution and supports continued radioecological monitoring of the catchment–lake system.

**Keywords:** Semipalatinsk Nuclear Test Site, Atomic Lake, radiocapacity, radionuclides, <sup>137</sup>Cs, <sup>241</sup>Am, <sup>152</sup>Eu, radioactive contamination

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## СЕМЕЙ ЯДРОЛЫҚ СЫНАҚ ПОЛИГОНЫНДАҒЫ АТОМ КӨЛІНІҢ РАДИОСЫЙЫМДЫЛЫҒЫН БАҒАЛАУ

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**Аннотация.** *Өзектілігі.* 1965 жылы Семей ядролық сынақ полигонында жүргізілген жерасты термоядролық жарылысы нәтижесінде пайда болған Атом көлі радиоактивті ластанған су нысаны болып табылады. Іргелес су жинау аумағында жинақталған радионуклидтер қайта таралып, жер үсті ағынымен көлге түсуі мүмкін, сондықтан олардың ұзақ мерзімді динамикасын бағалау қажет. *Мақсаты.* Зерттеудің мақсаты - радиоактивті ыдырауды ескере отырып, 2014–2020 жылдар аралығындағы су жинау аумақтарындағы  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  және  $^{152}\text{Eu}$  динамикасын талдау негізінде ішкі және сыртқы Атом көлдерінің радиосыйымдылығының өзгеруін бағалау. *Әдістері.* Далалық және зертханалық гамма-спектрометрия әдістерімен алынған 2014 және 2020 жылдардағы мониторинг деректері талданды. Су жинау аумақтарының шекаралары QGIS көмегімен анықталды. Радионуклидтердің жиынтық белсенділігі меншікті белсенділік мәндері, ластанған секторлар саны және топырақ тығыздығының параметрлері бойынша есептелді. Уақыт бойынша өзгерістер радионуклидтердің жартылай ыдырау кезеңдерін ескере отырып бағаланды. *Нәтижелері.* Ішкі көлдің есептік су жинау ауданы 0,16 км<sup>2</sup>, сыртқы көлдікі 3,07 км<sup>2</sup> құрады. 2014–2020 жылдары  $^{137}\text{Cs}$  есептік жиынтық белсенділігі  $2,169 \times 10^9$ -нан  $1,158 \times 10^9$  Бк-ке,  $^{241}\text{Am}$  -  $2,906 \times 10^8$ -ден  $2,663 \times 10^8$  Бк-ке, ал  $^{152}\text{Eu}$  -  $1,018 \times 10^9$ -нан  $5,288 \times 10^8$  Бк-ке дейін төмендеді. Радиоактивті ыдырауды ескергеннен кейінгі есептеулер зерттелген су жинау аумақтарында радионуклидтердің қайта таралуы мен мөлшерінің азаюын көрсетті. Өзгерістер жекелеген радионуклидтер мен көл жүйесінің ішкі және сыртқы бөліктері арасында ерекшеленді. *Қорытындылар.* Нәтижелер 2014–2020 жылдары су жинау аумағы–Атом көлі жүйесіндегі радионуклидтер қорының өзгергенін және бұл өзгерістерді тек радиоактивті ыдыраумен түсіндіру жеткіліксіз екенін көрсетті. Ұсынылған тәсіл радионуклидтердің ұзақ мерзімді қайта таралуын бағалауға мүмкіндік береді және жүйелі радиоэкологиялық мониторингті жалғастыру қажеттілігін негіздейді.

**Түйін сөздер:** Семей ядролық сынақ полигоны, Атом көлі, радиосыйымдылық, радионуклидтер,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{152}\text{Eu}$ , радиоактивті ластану

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## ОЦЕНКА РАДИОЕМКОСТИ АТОМНОГО ОЗЕРА НА СЕМИПАЛАТИНСКОМ ЯДЕРНОМ ПОЛИГОНЕ

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**Аннотация.** *Актуальность.* Атомное озеро, образовавшееся в результате подземного термоядерного взрыва на Семипалатинском испытательном ядерном полигоне в 1965 году, остается радиоактивно загрязненным водным объектом. Радионуклиды, накопленные на прилегающей водосборной территории, могут перераспределяться и поступать в озеро с поверхностным стоком, что обуславливает необходимость оценки долгосрочной динамики их содержания. *Цель.* Оценить изменение радиоемкости внутреннего и внешнего Атомных озер на основе анализа динамики содержания радионуклидов  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  и  $^{152}\text{Eu}$  на их водосборных территориях в период с 2014 по 2020 год с учетом процессов радиоактивного распада. *Методы.* Проанализированы данные мониторинга за 2014 и 2020 годы, полученные с использованием методов полевой и лабораторной гамма-спектрометрии. Границы водосборных территорий определены с помощью программного обеспечения QGIS. Суммарную активность радионуклидов рассчитывали на основе значений удельной активности, количества загрязненных секторов и плотности почвы. Динамику активности радионуклидов оценивали с учетом

периодов их полураспада. *Результаты.* Расчетная площадь водосбора внутреннего озера составила 0,16 км<sup>2</sup>, внешнего — 3,07 км<sup>2</sup>. В период с 2014 по 2020 год расчетная суммарная активность <sup>137</sup>Cs уменьшилась с  $2,169 \times 10^9$  до  $1,158 \times 10^9$  Бк, <sup>241</sup>Am — с  $2,906 \times 10^8$  до  $2,663 \times 10^8$  Бк, <sup>152</sup>Eu — с  $1,018 \times 10^9$  до  $5,288 \times 10^8$  Бк. Расчеты с учетом радиоактивного распада выявили изменения запасов радионуклидов на исследованных водосборных территориях, указывающие на их перераспределение и снижение суммарной активности. При этом установлены различия в динамике отдельных радионуклидов между внутренней и внешней частями озерной системы. *Выводы.* Результаты исследования свидетельствуют об изменении запасов радионуклидов в системе «водосбор — Атомное озеро» в период с 2014 по 2020 год, которое не может быть объяснено исключительно процессами радиоактивного распада. Предложенный методический подход позволяет оценивать долгосрочное перераспределение радионуклидов и подтверждает необходимость продолжения радиоэкологического мониторинга системы «водосбор — озеро».

**Ключевые слова:** Семипалатинский испытательный ядерный полигон, Атомное озеро, радиоемкость, радионуклиды, <sup>137</sup>Cs, <sup>241</sup>Am, <sup>152</sup>Eu, радиоактивное загрязнение.

**Introduction.** Radioactive contamination of surface waters remains an important environmental safety issue associated with nuclear weapons testing, radiation accidents, and nuclear industry activities. One of the most significant radioactively contaminated territories is the Semipalatinsk Nuclear Test Site (SNTS) in northeastern Kazakhstan, where 456 nuclear tests, including 116 atmospheric and 340 underground explosions, were conducted between 1949 and 1989. Although the SNTS was closed in 1991, long-term radioecological monitoring has demonstrated the persistence and migration of radionuclides in its ecosystems (Nazarbayev et al., 2016).

On January 15, 1965, an underground thermonuclear explosion at a depth of 178 m at the Balapan test site formed the Atomic Lake (AL) in the floodplain of the Chagan River. The explosion created a crater approximately 430 m in diameter and 100 m deep. The resulting lake system consists of an inner reservoir (0.14 км<sup>2</sup>; approximately 7 million м<sup>3</sup>) and an outer reservoir (3.5 км<sup>2</sup>; approximately 10 million м<sup>3</sup>) (Subbotin et al., 2020). The ring-shaped elevations surrounding the inner lake restrict direct surface runoff, whereas the outer lake receives runoff from a considerably larger catchment area.

Radionuclides deposited within the surrounding territory may be transported into the lake system by stormwater and snowmelt runoff, groundwater discharge, and other hydrological processes (Aidarkhanov et al., 2013; Larionova et al., 2017; Razakova et al., 2020; Aidarkhanova et al., 2022). Previous studies have demonstrated the presence and redistribution of <sup>3</sup>H, <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>152</sup>Eu, and <sup>239+240</sup>Pu in the Atomic Lake and surrounding environment (Ochiai et al., 2013;

Aidarkhanova et al., 2018). Radionuclide migration into aquatic systems depends on hydrological conditions, soil properties, slope characteristics, precipitation, radioactive decay, and sorption processes (Parker et al., 1966; Fabre et al., 1980; Yabusaki et al., 2017).

Secondary radionuclide input from contaminated catchments may therefore contribute to the long-term radiocapacity of the Atomic Lake system. This process is particularly important because radionuclides entering a water body can subsequently accumulate and redistribute among water, suspended matter, and bottom sediments. Previous investigations of the Atomic Lake and other contaminated lake systems at the Semipalatinsk Test Site have mainly focused on radionuclide concentrations and migration in individual environmental components (Aktayev et al., 2025), while quantitative changes in radionuclide inventories within the catchment–lake system remain insufficiently characterized. Figure 1 shows a picture of the inner and outer Atomic Lakes.



Figure 1. The inner and outer Atomic Lakes.

The general dynamics of radionuclides are presented at the diagram which shows that the radioactive capacity of the lake should increase over the time (Fig.2).

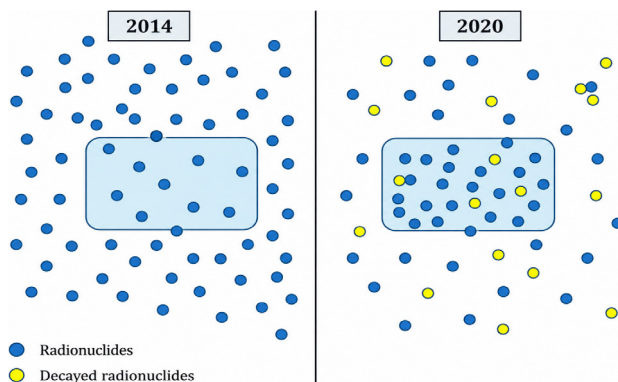


Figure 2. Dynamics of the distribution of radionuclides in the catchment area of the AL.

The hypothesis of this study is that secondary radionuclide contamination of the Atomic Lake (AL) is associated with the leaching and transport of radionuclides from contaminated surface soils of the catchment area by precipitation and surface runoff. However, the contribution of radionuclide input from the catchment area to the radiocapacity of the AL has not previously been quantitatively assessed.

**Materials and Methods.** The study objects were the inner and outer Atomic Lakes and their adjacent catchment areas at the Balapan site of the Semipalatinsk Nuclear Test Site. The inner lake is a rounded explosion crater connected by a narrow channel to the outer lake, which extends along the Chagan River floodplain (Fig. 3).

The study area has a sharply continental climate characterized by hot, moderately dry summers and cold winters. January temperatures range from  $-27$  to  $-33$  °C, while maximum July temperatures reach  $+32$  to  $+37$  °C. Average monthly wind speeds generally range from 2.9 to 3.6 m/s, with the strongest winds occurring mainly in spring (Kakimov et al., 2013).

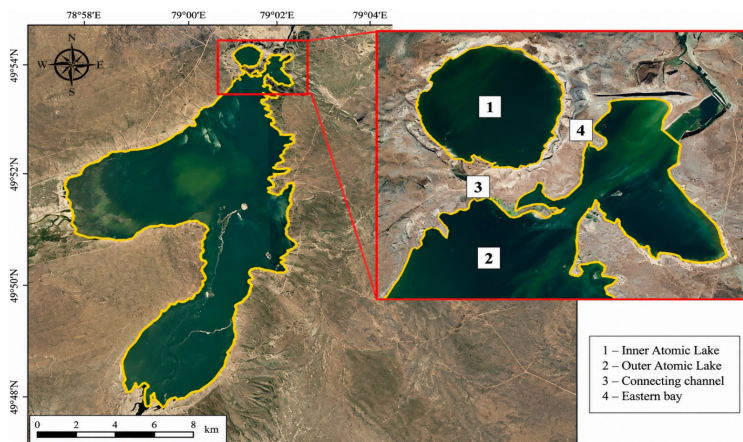


Figure 3. Location of the lakes' system: (1) The inner Atomic lake; (2) The outer Atomic lake; (3) The channel connecting the inner and the outer Atomic lakes.

Determination of the catchment area and total radionuclide activity. The catchment areas of the inner and outer Atomic Lakes were delineated in QGIS 3.38 and divided into  $100 \times 100$  m sectors ( $10,000 \text{ m}^2$  or  $0.01 \text{ km}^2$ ) (Fig. 4). The inner lake is surrounded by ring-shaped elevations formed by the explosion, which restrict surface runoff into the crater. Consequently, most surface runoff is directed toward the outer lake, whereas only precipitation and runoff originating within the ring-shaped elevation contribute to the inner lake catchment. Catchment areas were calculated using the \$area tool in QGIS.

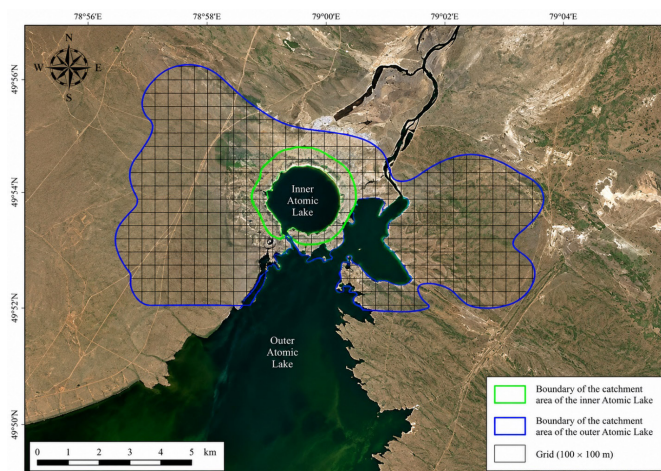


Figure 4. The boundaries of the catchment area of the inner and the outer Atomic lakes.

Spatial distribution maps of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  were constructed using the results of field gamma-spectrometric surveys conducted in 2014 and 2020 (Figs. 5–7). These maps were subsequently used to estimate the total radionuclide inventories within the delineated catchment areas.

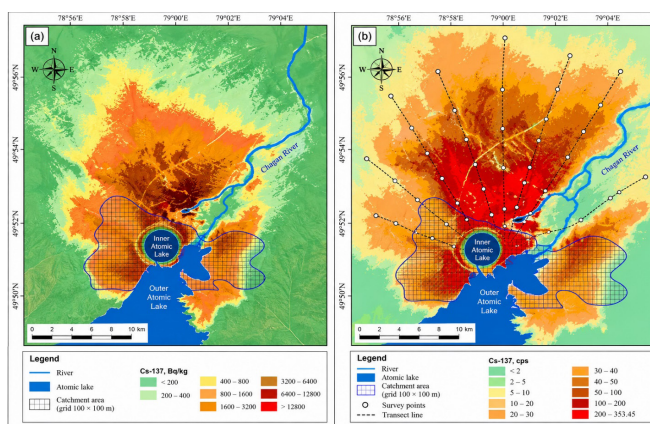


Figure 5.  $^{137}\text{Cs}$  distribution area by its concentration in 2014 (a) and 2020 (b).

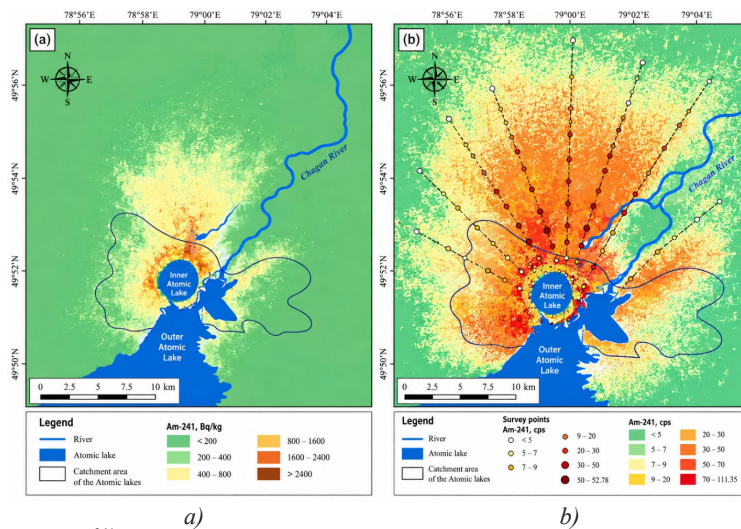


Figure 6.  $^{241}\text{Am}$  distribution area by its concentration in 2014 (a) and 2020 (b).

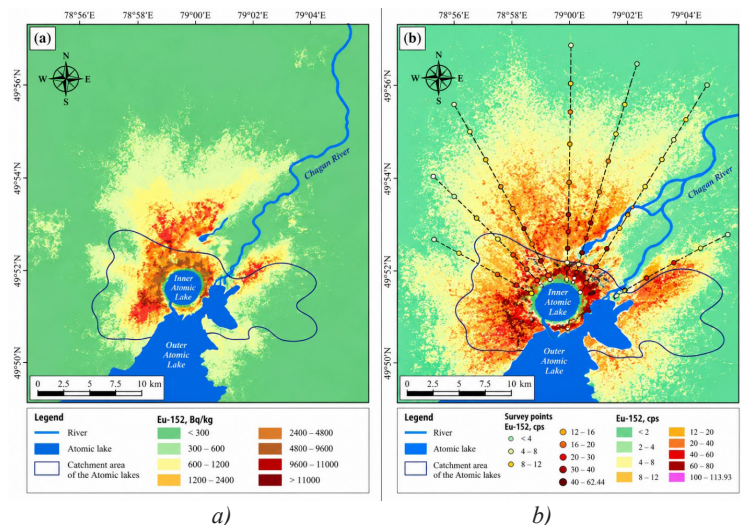


Figure 7.  $^{152}\text{Eu}$  distribution area by its concentration in 2014 (a) and 2020 (b).

Determination of total radionuclide activity in 2014. The total activities of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  within the catchment areas were estimated using the spatial contamination data presented in Figures 5–7. For each activity interval, its upper limit was used in the calculations. For example, the  $^{137}\text{Cs}$  intervals were represented by 200, 400, 800, 1600, 3200, 6400, and 12,800 Bq/kg, with a separate category for values >12,800 Bq/kg. The areas corresponding to each contamination interval were determined from the maps and summarized in Table 1. Total radionuclide activity was calculated as follows:

$$V = A \times n, \quad (1)$$

where  $A$  is the specific activity of a particular sector, Bq/kg

$n$  is the number of sectors with the same specific activity.

Thus, there are 16 sectors with a specific activity of 200 Bq/kg.

It was considered that the specific gravity of the soil is 1450 kg/m<sup>3</sup>. As a result, the total radioactivity of the isotope in the entire catchment area was determined using the following formula:

$$R_I = V \times M_{\text{soil}} \quad (2)$$

where  $R_I$  is the total radioactivity of the isotope, Bq/kg

$V$  is the number of sectors with the same specific activity,

$M_{\text{soil}}$  is the specific gravity of the soil, 1450 kg/m<sup>3</sup>.

The work used field gamma-spectrometric systems based on scintillation detectors with lanthanum bromide (LaBr<sub>3</sub>(Ce)) crystals and ORTEC DigiBase pulse analyzers.

**Pedestrian gamma-spectrometric survey.** The survey was conducted along predetermined profiles spaced 20 m apart using satellite navigation. The operator moved at a constant speed of 1–2 km/h, with the detector fixed at a height of 0.5 m above the ground. Gamma spectra were continuously recorded at 10-s intervals and stored on a mobile computer, yielding approximately 1,800 spectra per survey sector.

Approximately 600,000 gamma spectra were obtained and automatically processed using the *Spectrtool* software to generate data for mapping surface radioactive contamination. The detection sensitivity was 100 Bq/kg for <sup>137</sup>Cs, 200 Bq/kg for <sup>241</sup>Am, and 300 Bq/kg for <sup>152</sup>Eu.

**Determination of total radionuclide activity in 2020.** Field measurements were performed using an ORTEC scintillation gamma spectrometer equipped with a LaBr<sub>3</sub> detector. Gamma spectra were acquired for 300 s at predetermined measurement points, with the detector positioned 0.5 m above the ground and at an angle not exceeding 30° from the vertical axis. The spectra and corresponding geographic coordinates were automatically synchronized and recorded on a portable computer.

The obtained data were processed using Golden Software Surfer and ArcGIS to construct spatial distribution maps of artificial gamma-emitting radionuclides. At 50 measurement points, soil samples were collected from the upper 5-cm layer for laboratory gamma-spectrometric analysis. Measurements were performed using an ORTEC gamma-spectrometric system equipped with a 1.5-inch LaBr<sub>3</sub> scintillation detector. The detector was selected because of its thermal stability and energy resolution, allowing identification of the characteristic gamma lines of <sup>241</sup>Am (59.5 keV), <sup>152</sup>Eu (121 keV), and <sup>137</sup>Cs (662 keV).

Calibration was performed to establish conversion factors between the field detector count rate (cps) and radionuclide-specific activity in soil (Bq/kg). Based on the calibration results, correlations between the measured count rate and specific activity were determined for each radionuclide. For example, for <sup>137</sup>Cs:

$$A \text{ (Bq/kg)} = 71.284 \times X \text{ (pps)}, \quad (3)$$

Thus, the total radioactivity of an isotope is determined by the following formula:

$$R_2 = A \times n \times M_{\text{soil}}, \quad (4)$$

where  $R_2$  is the total radioactivity of the isotope, Bq/kg  
 $n$  is the number of sectors with the same specific activity  
 $M_{\text{soil}}$  is the specific gravity of the soil, 1450 kg/m<sup>3</sup>.

*The method of determining the radio capacity of the Atomic lakes.* Background monitoring data obtained by the National Nuclear Center of the Republic of Kazakhstan in 2014 and 2020 were used as the initial data for determining the radiological capacity of the Atomic Lake (Serzhanova et al., 2018; Kadyrzhanov et al., 2000). To determine the radiological capacity of the Atomic Lake, the change in total radionuclide activity between 2014 and 2020 was calculated, taking into account radioactive decay over the corresponding period.

The amount of radionuclide activity removed from the catchment area was estimated from the difference between the radionuclide inventories measured in 2014 and 2020. The expected decrease in activity due to radioactive decay over the seven-year period was taken into account in order to distinguish radioactive decay from radionuclide removal from the catchment area.

The decrease in total radionuclide activity due to radioactive decay over the seven-year period was calculated using the following equation (5):

$$X = R_1 - R_2, \quad (5)$$

where  $X$  is the volume of radiation, Bq/kg

$R_1$  is the total radioactivity in the territory of the Atomic lake in 2014, Bq/kg

$R_2$  is the total radioactivity in the territory of the Atomic lake in 2020, Bq/kg.

*Determination of the decay rate of isotopes*

The decay rate of isotopes in soil and water of Atomic lakes was calculated based on the law of radioactive decay and determined by the formulas (6, 7).

$$a = N_0 e^{-\lambda t}, \quad (6)$$

where  $N_0$  is the initial activity of radionuclides, i.e. the number of atoms for  $t=0$ , Bq/kg

$\lambda$  is the decay constant, which characterizes the probability of radioactive decay per unit time and has the dimension of a  $c^{-1}$ ,

$t$  is the period of time during which the radionuclide loses half of its radioactivity, years.

$$\lambda = \frac{\ln 2}{dt}, \quad (7)$$

The negative of the decay constant indicates the decrease in the number of radioactive atoms over the time. The half-life for  $^{137}\text{Cs}$  – 30,1 years, for  $^{241}\text{Am}$  – 432,2 years;  $^{152}\text{Eu}$  - 13,5 years (Muller, 2010).

The half-life of radionuclides was subtracted from the difference between radiation activity in 2014 and 2020 for 7 years and the amount of radiation was obtained considering the decay of radioisotopes from the catchment area (Z) which was washed away into Atomic lakes by the formula (8).

$$Z = X - a, \quad (8)$$

where Z is the decay of radioisotopes,

X is the total radioactivity in the territory of the Atomic lake in 2014, Bq/kg

a is the total radioactivity in the territory of the Atomic lake in 2020, Bq/kg.

Modern radio capacity (A) of the Atomic lakes characterized by a combination of radiation that was washed off the catchment area considering the decay of radioisotopes (Z) as well as the activity of radionuclides considering their decay (Y) in the water itself. It is calculated using the following formula (9).

$$A = Z + Y, \quad (9)$$

where A is the modern radio capacity,

Z is the decay of radioisotopes,

Y is the decay (Y) in the water.

The calculations for determining the radio capacity of the Atomic lakes were conducted for the inner and the outer Atomic lakes separately, in order to determine the migration and redistribution of the radionuclides.

**Results.** Characteristics of the Atomic Lakes and their catchment areas. The mapped area of the inner and outer Atomic Lakes was 16.32 km<sup>2</sup>, comprising 1.62 km<sup>2</sup> for the inner lake, 14.19 km<sup>2</sup> for the outer lake, and 0.51 km<sup>2</sup> for the connecting channel. QGIS analysis gave catchment areas of 0.16 km<sup>2</sup> for the inner lake and 3.07 km<sup>2</sup> for the outer lake.

Changes in radionuclide inventories between 2014 and 2020. Total activities of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  were calculated for the 2014 and 2020 monitoring datasets. For each mapped activity class, total activity was obtained from the specific activity, the number of contaminated sectors, and the soil mass represented by those sectors. The same calculation procedure was applied to all three radionuclides. To avoid repeating sector-by-sector calculations, the resulting inventories are summarized below.

Table 1. Radionuclide inventories in the catchment soils and lake water in 2014 and 2020.

| Radionuclide      | Lake  | Soil 2014, Bq | Soil 2020, Bq | Water 2014, Bq | Water 2020, Bq |
|-------------------|-------|---------------|---------------|----------------|----------------|
| <sup>137</sup> Cs | Outer | 1,732,460,000 | 790,614,408   | 122,090,204    | 55,716,308     |
| <sup>137</sup> Cs | Inner | 436,160,000   | 367,244,475   | 269,234,568    | 226,694,121    |
| <sup>241</sup> Am | Outer | 261,580,000   | 245,065,453   | 18,434,109     | 17,270,293     |
| <sup>241</sup> Am | Inner | 29,000,000    | 21,283,100    | 17,901,235     | 13,137,716     |
| <sup>152</sup> Eu | Outer | 726,160,000   | 419,565,852   | 51,174,066     | 29,567,713     |
| <sup>152</sup> Eu | Inner | 292,320,000   | 109,244,160   | 180,444,444    | 67,434,667     |

In the outer-lake catchment, the soil inventory decreased most strongly for <sup>137</sup>Cs, from  $1.73 \times 10^9$  Bq in 2014 to  $7.91 \times 10^8$  Bq in 2020. The <sup>152</sup>Eu inventory also decreased markedly, from  $7.26 \times 10^8$  to  $4.20 \times 10^8$  Bq, whereas <sup>241</sup>Am changed comparatively little ( $2.62 \times 10^8$  to  $2.45 \times 10^8$  Bq). In the inner-lake catchment, the largest relative reduction was observed for <sup>152</sup>Eu ( $2.92 \times 10^8$  to  $1.09 \times 10^8$  Bq).

For the lake-water estimates, <sup>137</sup>Cs remained the largest inventory in the outer lake, decreasing from  $1.22 \times 10^8$  to  $5.57 \times 10^7$  Bq. In the inner lake, the <sup>152</sup>Eu estimate decreased from  $1.80 \times 10^8$  to  $6.74 \times 10^7$  Bq, while <sup>137</sup>Cs decreased from  $2.69 \times 10^8$  to  $2.27 \times 10^8$  Bq.

#### Activity loss and radioactive decay

The observed decrease between 2014 and 2020 includes both radionuclide redistribution/removal from the catchment and radioactive decay. The decay-related component was therefore evaluated separately using the radionuclide half-lives. Table 2 presents the observed activity loss (X) and the activity component attributed to radioactive decay (Z) for the outer and inner systems.

Table 2. Observed activity loss (X) and decay-related activity component (Z) between 2014 and 2020.

| Medium | Radionuclide      | X, outer lake, Bq | X, inner lake, Bq | Z, outer lake, Bq | Z, inner lake, Bq |
|--------|-------------------|-------------------|-------------------|-------------------|-------------------|
| Soil   | <sup>137</sup> Cs | 941,845,592       | 68,915,525        | 683,924,513       | 3,981,929         |
| Soil   | <sup>241</sup> Am | 16,514,547        | 7,716,900         | 13,594,379        | 7,393,156         |
| Soil   | <sup>152</sup> Eu | 306,594,148       | 183,075,840       | 87,357,964        | 94,821,018        |
| Water  | <sup>137</sup> Cs | 66,373,897        | 42,540,447        | 48,197,640        | 2,457,981         |
| Water  | <sup>241</sup> Am | 1,163,816         | 4,763,519         | 958,025           | 4,563,677         |
| Water  | <sup>152</sup> Eu | 21,606,353        | 113,009,778       | 6,156,305         | 58,531,493        |

The largest observed soil-activity loss occurred for <sup>137</sup>Cs in the outer-lake catchment ( $9.42 \times 10^8$  Bq), followed by <sup>152</sup>Eu ( $3.07 \times 10^8$  Bq). For <sup>241</sup>Am, the corresponding loss was much smaller ( $1.65 \times 10^7$  Bq). In the inner system, the largest soil-activity loss was observed for <sup>152</sup>Eu ( $1.83 \times 10^8$  Bq). These differences indicate that the temporal changes cannot be described by radioactive decay alone.

#### Estimated modern radio capacity

Using the activity balance adopted in this study, the modern radio capacity was estimated separately for the outer and inner Atomic Lakes.

Table 3. Estimated modern radio capacity of the Atomic Lakes.

| Radionuclide      | Outer Atomic Lake, Bq | Inner Atomic Lake, Bq |
|-------------------|-----------------------|-----------------------|
| $^{137}\text{Cs}$ | 732,122,153           | 6,439,909             |
| $^{241}\text{Am}$ | 14,552,404            | 11,956,833            |
| $^{152}\text{Eu}$ | 93,514,268            | 153,352,511           |

The outer Atomic Lake was dominated by  $^{137}\text{Cs}$ , with an estimated radio capacity of  $7.32 \times 10^8$  Bq, while  $^{152}\text{Eu}$  contributed  $9.35 \times 10^7$  Bq and  $^{241}\text{Am}$   $1.46 \times 10^7$  Bq. In the inner Atomic Lake,  $^{152}\text{Eu}$  was the largest component ( $1.53 \times 10^8$  Bq), whereas the estimated contributions of  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  were  $1.20 \times 10^7$  and  $6.44 \times 10^6$  Bq, respectively.

Overall, the 2014–2020 comparison shows radionuclide-specific redistribution between the catchment soils and lake system. The pronounced changes in  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$ , together with the smaller changes in  $^{241}\text{Am}$ , are consistent with the combined influence of radioactive decay and transport from the catchment. The present dataset supports quantification of these inventory changes; interpretation of retention in bottom sediments should be discussed separately where sediment measurements are available.

Figures 8-9 show the dynamics of radiation decay, considering the decay in the soil and water of the Atomic lake.

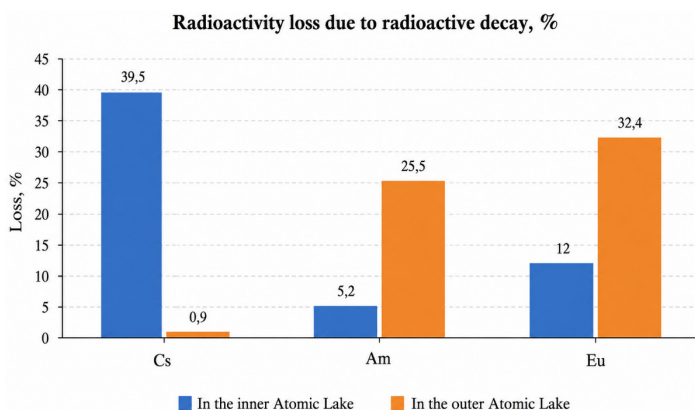


Figure 8. The amount of radiation loss in the soil considering the decay in the inner and outer Atomic lakes.

It can be seen that the concentration of  $^{152}\text{Eu}$  in the catchment area has decreased over the period 2014–2020, but remains high in several sectors, especially in the inner Atomic lake's catchment area.

The increase in  $^{152}\text{Eu}$  activity in some areas indicates the local accumulation of radionuclides, either due to re-distribution or delayed leaching.

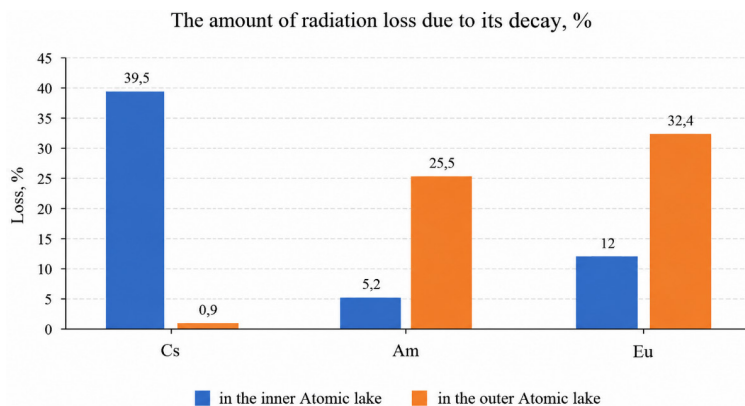


Figure 9. amount of radiation loss in the water considering the decay in the inner and outer Atomic lakes.

**Discussion.** The results demonstrate substantial changes in the calculated inventories of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  within the catchment areas of the inner and outer Atomic Lakes between 2014 and 2020. After correction for radioactive decay, the estimated losses from the catchment areas amounted to 738.6 MBq for  $^{137}\text{Cs}$ , 26.5 MBq for  $^{241}\text{Am}$ , and 246.9 MBq for  $^{152}\text{Eu}$ . These differences indicate radionuclide-specific behavior and suggest that radioactive decay alone cannot explain the observed changes.

The decrease in  $^{137}\text{Cs}$  was the most pronounced. Its redistribution may be associated with surface runoff and transport with suspended soil particles, consistent with previous studies demonstrating the mobility of radiocesium in surface-water systems (Strakhovenko et al., 2010). In contrast,  $^{241}\text{Am}$  showed the smallest decrease, which is consistent with its long half-life (432.2 years) and stronger association with soil and sediment particles. The behavior of  $^{152}\text{Eu}$  reflects both environmental redistribution and its considerably shorter half-life (13.5 years), which accounts for a substantial fraction of its activity decrease during the study period.

Hydrological variability is an important factor controlling radionuclide redistribution within the Atomic Lake system. Seasonal flooding, precipitation, surface runoff, changes in water level, and sedimentation can influence the transfer of radionuclides between catchment soils, water, suspended matter, and bottom sediments. (Umirbayeva, 2022) similarly associated variations in technogenic radionuclide concentrations in the Atomic Lake system with instability of the hydrological regime and possible remobilization from contaminated soils and sediments.

The spatial distributions of radionuclides (Figs. 5–7) indicate that contamination remains concentrated around the Atomic Lake and within its catchment areas. The geomorphological difference between the inner and outer lakes is particularly important. The inner lake is surrounded by explosion-generated elevations that

restrict surface runoff, whereas the outer lake receives runoff from a considerably larger area. Consequently, radionuclide transfer from surrounding soils is expected to differ between the two catchments.

Groundwater transport should be interpreted separately from surface runoff. Previous investigations of the geological and lithological conditions at the Balapan site identified low-permeability clays, clay shales, and siltstones that can restrict groundwater migration from underground nuclear-test areas. (Aktayev et al. 2025) reported that groundwater originating from the Balapan test site did not significantly affect tritium contamination of the Atomic Lake. Therefore, the present results do not provide sufficient evidence to identify groundwater as the dominant pathway for the radionuclides investigated here.

Overall, the observed changes support the hypothesis that radionuclides deposited in the catchment are progressively redistributed within the catchment–lake system. Surface runoff, erosion and particle transport, sorption–desorption processes, sedimentation, and radioactive decay may contribute simultaneously. However, the present inventory-based approach cannot quantify the individual contribution of each mechanism. Direct measurements of radionuclide fluxes in runoff, lake water, suspended matter, and bottom sediments are therefore required to establish a complete radionuclide mass balance.

**Conclusions.** This study quantified changes in the inventories of  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and  $^{152}\text{Eu}$  within the catchment areas of the inner and outer Atomic Lakes between 2014 and 2020. The calculated catchment areas were 0.16 km<sup>2</sup> for the inner lake and 3.07 km<sup>2</sup> for the outer lake.

Between 2014 and 2020, the calculated total activity within the investigated catchment areas decreased from  $2.169 \times 10^9$  to  $1.158 \times 10^9$  Bq for  $^{137}\text{Cs}$ , from  $2.906 \times 10^8$  to  $2.663 \times 10^8$  Bq for  $^{241}\text{Am}$ , and from  $1.018 \times 10^9$  to  $5.288 \times 10^8$  Bq for  $^{152}\text{Eu}$ . After correction for radioactive decay, the estimated activity losses from the catchment areas were 738.6 MBq for  $^{137}\text{Cs}$ , 26.5 MBq for  $^{241}\text{Am}$ , and 246.9 MBq for  $^{152}\text{Eu}$ .

The largest calculated loss was observed for  $^{137}\text{Cs}$ , whereas  $^{241}\text{Am}$  showed the smallest change. These radionuclide-specific differences indicate that radioactive decay alone cannot explain the observed temporal changes and that environmental redistribution within the catchment–lake system contributes to the evolution of radionuclide inventories.

The results support the occurrence of secondary radionuclide transfer from contaminated catchment soils toward the Atomic Lake system. However, the present approach estimates changes in radionuclide inventories and does not directly quantify their accumulation in bottom sediments or distinguish the individual contributions of surface runoff, groundwater transport, erosion, and other migration pathways.

Further research should therefore focus on simultaneous monitoring of catchment soils, surface runoff, lake water, suspended matter, and bottom sediments,

combined with seasonal hydrological observations. Such measurements will allow a quantitative radionuclide mass balance to be established and will improve long-term predictions of radionuclide redistribution and the radioecological state of the Atomic Lake system.

*Data Availability Statement: The data supporting the findings of this study are available from the bulletins of the National Nuclear Center of the Republic of Kazakhstan. For further details on accessing the dataset, researchers may contact the corresponding author.*

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