INSINUME 2019
8th International Symposium on IN Situ NUclear MEtrology as a tool for radioecology
23-26 April 2019
Richmond Ephesus Resort, KUSADASI-TURKEY

FULL TEXT BOOK

- Regulatory framework concerning the release of radionuclides and the monitoring of radioactivity in the environment
- New technologies and methodologies in radioactivity metrology
- Quality Assurance
- Radioactivity levels in environment and transfer of radionuclides
- Impact of radioactivity on environment, population and biota
- Nuclear metrology in incidental/accidental conditions
8th INTERNATIONAL SYMPOSIUM ON “IN SITU NUCLEAR METROLOGY AS A TOOL FOR RADIOECOLOGY”

INSINUME 2019

FULL TEXT BOOK

Publication of the Full Texts issued in this book is possible provided that written approval of the author and editor are taken.


Editors
Sabriye YUŞAN
Şule AYTAŞ

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PROGRAMME

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Symposium Venue:
Richmond Ephesus Resort Hotel
Kuşadası, TURKEY
23 APRIL 2019, TUESDAY

08.30-10.30 ORAL PRESENTATIONS - HALL A

10.30-10.45 IN SITU NORM BIOMONITORING SYSTEMS IN INDUSTRIAL DEPOSITS
Anita Coiard, Anita Peka, Edit Toth-Bodrog, Tibor Kovacs

10.45-11.00 CURRENT STATE OF THE HUNGARIAN INDOOR RADON MAP
Tibor Kovacs, Anikó Kocsis, Anita Coiard, Katalin Zsuzsanna Szabo, Zoltan Nemeth

11.00-11.15 THE CORRELATION OF THE SEISMIC ACTIVITIES AND RADON CONCENTRATION IN GROUNDWATER
Mihály Békei, Szub

11.15-11.30 RESEARCHING GENETIC EFFECTS OF CHRONIC IRRADIATION OF \nJUDE GRASS POPULATION GROWING IN VENUES OF TESTING \nWARFARE RADIOACTIVE AGENTS
Karoly Miltnerova, Jozef Bojplansky

11.30-11.45 INFLUENCE OF CONTAMINATED SEDIMENTS IN SICHENKOV
RESERVOIR ON 40K CONCENTRATION IN FISH
Suzanna Kezmanowska, Maxim Ivanov, Toshiharu WADA, Koji MAEDA, \nLudmila ESHTOVA, Antoniy GUDOV, Nadezhda VADILKOVA, Evgeny \nKONSTANTINOV, Alexandru ROZKOVAY, Valentin GOLOSOV

11.45-12.00 MEASUREMENT AND EVALUATION OF RADIATION EXPOSURE AND \nRISKS TO THE PUBLIC IN THE URANIUM AND THORIUM MINES AREAS
Yukinori Akaoka

12.00-13.30 LUNCH

13.30-14.00 FEEDBACK ON THE BELGIAN IMPLEMENTATION OF THE 2015/67 \nECRD IN DRINKING WATER DIRECTIVE - THE TECHNICAL CHALLENGES \nFOR AN ENVIRONMENTAL AQUACULTURE
Chairman: Dr. Michel BERNOICH, NDR

14.00-14.15 PRELIMINARY STUDY OF COMPTON SUPPRESSION SYSTEM FOR \nIN SITU MONITORING NUCLEAR DECOMMISSIONING SITES
Chamiki Lee, Hee Reung Kim

14.15-14.30 REMOVAL OF RADIONUCLIDES FROM AQUEOUS SOLUTIONS BY \nSORPTION TECHNIQUES
Fathal Noil

14.30-14.45 ARTIFICIAL RADIONUCLIDES MIGRATION PATHWAYS IN \nRESERVOIRS R-17 AND R-4 (PLANT MAYAK)
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14.45-15.00 DETERMINING THE 87/240 + 207/209 FISSION PRODUCTS RATIO AT THE \n(Experimental Field) Site
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15.00-15.15 COFFEE BREAK

15.15-16.15 ORAL PRESENTATIONS - HALL A

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Gilles Osseyan

15.30-15.45 A CASE STUDY: RS-222 IN RAIN WATER A FINGERPRINT FOR \nEARTHQUAKE?
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15.45-16.00 EARTHWORM SPECIES AS IN INDIVIDUAL ORGANOISOMS \nNORM TRANSFER IN SOIL ECOSYSTEMS
Domiokos Madar, Ferenc Buday, Anita Peka, Edit Toth-Bodrog, Tibor \nKovacs

16.00-16.15 ARTIFICIAL AND NATURAL RADIONUCLIDES IN LAKES OF CRIMEA
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16.15-16.30 BREAK

16.30-17.45 ORAL PRESENTATIONS - HALL A

16.30-16.45 BIOMONITORING OF RADIOACTIVE CONTAMINATION OF THE \nYENISEI RIVER
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16.45-17.00 PLUTONIUM AND AMERICIUM IN THE DEEP BLACK SEA BOTTOM \nSEDIMENTS
Vladislav Yu. Prokoch, Natalia N. Tershevchko, Artem A. Papiskiv, \nOlga D. Chumachenko-Prokochina

17.00-17.15 DYNAMICS OF DOSE EQUIVALENT RATE ABOVE THE SOIL \SURFACE AS INDICATOR OF NATURAL ATTENUATION PROCESSES
Alexey Kompanev, Toshihiko Yoshinari, Yoshifumi Makajima

17.15-17.30 MCNP MODELING OF REAL-TIME AIRBORNE ALPHA \BETA \DETECTION SYSTEM
Si Hyung Sung, Hee Reung Kim

17.30-18.00 CANBERRA PRESENTATION
**8th International Symposium on IN Situ NUclear MEtrology as a tool for radioecology**

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P44. TECHNOGENIC RADIONUCLIDES DISTRIBUTION IN GRAIN-SIZE FRACTIONS OF SOIL IN THE AREA OF EXCAVATION NUCLEAR TEST (“ATOMIC LAKE”) Alisa Kadykova, Aysen Mendelev, Natalya Leonow

P45. DEVELOPMENT OF ACTIVITY RATIOS RESEARCH METHOD FOR THE 241Am AND 239Pu IN THE SOIL OF THE MAIN STUDY TEST SITES.
Vladislav Kudzuzh, Ildar Zvezdin, Anton Shatrev

HISTORICAL PLACES TO VISIT AT KUSADASI
Kusadasi is a beach resort town on Turkey’s western Aegean coast. A jumping-off point for visiting the classical ruins at nearby Ephesus (or Efes), it’s also a major cruise ship destination. Its seafarers promenade, marina, and harbor are lined with hotels and restaurants. Just offshore on Miletus Island is a walled Byzantine castle that once guarded the town, connected to the mainland via causeway.

EPHESEus ANCIENT CITY
Ephesus was an ancient Greek city on the coast of Ionia, three kilometres southwest of present-day Selçuk in Izmir Province, Turkey. It was built in the 10th century BC on the site of the former Arzawan capital by Attic and Ionian Greek colonists. Ephesus was an ancient port city whose well-preserved ruins are in modern-day Turkey. It was once considered the most important Greek city and the most important trading center in the Mediterranean region. Throughout history, Ephesus survived multiple attacks and changed hands many times between conquerors. It was also a hotbed of early Christian evangelism and remains an important archaeological site and Christian pilgrimage destination.

EPHESEUS MUSEUM
In Ephesus, the archeological artifacts that were dug up between 1867-1905 were transported to the British Museum; and findings from 1905-1923 were taken to Vienna. With the founding of the New Turkish Republic, the government forbade taking antiques out of the country and required the artifacts that were taken outside the country back to Turkey. In 1964, the Ephesus Museum was founded and the findings from the excavations at Ephesus archeological site and around were put to this museum. At this charming and well-organised museum, there are not only findings from the ongoing excavations at Ephesus archeological site, but also the artifacts from the Cukurçay Hound, the basilica of St. John, and the Temple of Artemis. At one of the sections, one can see a wide collection of coins dating back to when money was first used in history.

The museum of Ephesus, unlike most of other museums, is not designed according to the chronological order but the galleries are filled with artifacts according to a theme.

HOUSE OF VIRGIN MARY
House of Virgin Mary is located on the top of the “Bulbul” mountain 9 km ahead of Ephesus, the shrine of Virgin Mary enjoys a marvelous atmosphere hidden in the green. It is the place where Mary may have spent her last days. Indeed, she may have come in the area together with Saint John, who spent several years in the area to spread Christianity. Mary preferred this remote place rather than living in crowded place. The house of Virgin Mary is a typical Roman architectural example, entirely made of stones. In the 4th century AD, a church, combining her house and grave, has been built. The original two-storeyed house, which consisted of an ante-room (where today candles are proposed), bedroom and praying room (Christian church area) and a room with fireplace (chapel for Muslims). A front kitchen fell into ruins and has been restored in 1840’s. Today, only the central part and a room of the altar are open to visitors.
SİRİNCE VILLAGE
This pretty old Orthodox village, 12 km away from Ephesus and 30 km from Kusadasi, was once Cirklice (“ugly”), Indeed its habitants gave this name on purpose as they did not want to be bothered by foreigners nor to share the beauty of their village. Still, after years, visitors understood that the village was not ugly at all and called it Sıırınçe (“pretty”). As the village is located on the top of a mountain, anyone will enjoy the impressive vineyards’ and peach trees’ views on his way. Today the village is a perfect synthesis of Turk-Greek culture as of the 1920’s after the independence War, people exchange between Greek and Turks has occurred and all those typical Greek houses, though they kept their original outside characteristics, have received the local layout inside. The most beautiful specimens are open to visitors. And even in the courtyard one of them, one will discover a nicely restored Orthodox church.

GUVERCIHADA
PIGEON ISLAND
Guvercihada Fortress, located in the Hazaroschildar District of Kusadasi City, was built on the mouth of the harbour to protect it. There is an inner fortress (built by Barbaros Hayrettin Pasha) and fortress walls (built by Ilyas Aga) on the island. These walls were built to protect against the expected attacks from the Greek Islanders and from the sea during the Mora Revolt period. Walls were built approximately 3 m high and surrounded the island completely. The stones used in construction of the fortress were transported from Yalanci Cape. To the south of the walls, there is gate protected by two towers with arches and which can be climbed by means of stairs facing toward the south. While the north tower is in a pentagon shape, the southern tower is structured in a cylindrical form.

OKUZ MEHMET PASA KERVANSARAY
This caravansaray was built by the Grand Vizier Okuz Mehmed Pasha at the same time as the Kaleici Camii in Kusadasi to support maritime trade in the region. It is a brick construction, consisting of a two-storied portico around a courtyard approximately 18.5 meters (60 feet) long and 21.6 meters (71 feet) wide, where there was once a fountain. The main entrance is through a 2.96 m wide marble gate in the north. A second gate on the east side opens onto the market. Stairs are located in the northwest and southeast corners. The caravansaray was restored in 1965-1966 and converted to a hotel.

DILEK PENINSULA NATIONAL PARK
Dilek Peninsula National Park in Kusadasi (Dilek Peninsula – Great Menderes Delta National Park) is among the most important natural heritages of Turkey, with an area of 27,599 hectares. Dilek Peninsula, declared in 1966 as a national park, covers 29,985 hectares. Great Menderes Delta, declared in 1984 as national park, covers 16,615 hectares. The national park can be reached via the Kusadasi (Kusadasi) - Seke (Şıke) highway, and lies roughly in the middle of the two towns. It is possible to stay in the national park in tents or caravans, and there is food available. Dilek Peninsula National Park in Kusadasi has extraordinary beauty of Mediterranean flora with blue and emerald colored clean beaches.
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Institute for Radioelements – IRE and its subsidiary IRE ELIT
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Ege University
Ege University Institute of Nuclear Sciences
Preface

The 8th International Symposium on “IN SItu NUclear MEtrology as a tool for radioecology – INSINUME 2019 is organized by Ege University Institute of Nuclear Sciences and the Institute for Radioelements –IRE and subsidiary IRE ELIT, in cooperation with the International Atomic Energy Agency-IAEA between 23-26 April 2019 in Kuşadası-TURKEY

The previous INSINUME Symposia were organized in Ohrid (Macedonia 2017), Brussels (Belgium, 2012), Dubna (Russia, 2010), Rabat (Morocco, 2008), Kuşadası (Turkey, 2006), Albena (Bulgaria, 2004) and Fleurus (Belgium, 2002). The scope of the Symposium covers the following sessions:

- New technologies and methodologies in radioactivity metrology
- Nuclear metrology in incidental/accidental conditions
- Radioactivity levels in environment and transfer of radionuclides
- Impact of radioactivity on environment and population

The aim of the Symposium is to gather scientists, students, regulatory authorities, international organizations as well as industrial sector to exchange information and experience in the radiological monitoring of the environment and new technologies in nuclear metrology.

On behalf of Organizing Committee, we would like to thank the International Atomic Energy Agency-IAEA and other sponsors for providing financial support. We would like to express our appreciation to International Scientific Committee, Invited Speakers and participants who involved in INSINUME 2019 with their valuable contributions and efforts.

The selected papers will be published in a special Issue of The Journal of Environmental Radioactivity (Elsevier).

Yours Sincerely,

Sabriye YUŞAN
Ege University Institute of Nuclear Sciences
Izmir-TURKEY

Grégory DELECAUT
Institute of Radioelements
Fleurus-BELGIUM
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- Nuclear metrology in *incidental/accidental conditions*
- **Radioactivity levels** in environment and **transfer** of radionuclides
- **Impact of radioactivity** on environment and biota
RADIONUCLIDIC IMPURITY DETERMINATION OF $^{99m}$Tc AND EVALUATION OF UNCERTAINTY BUDGET

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ABSTRACT

Radionuclides are essential for nuclear medicine, especially for imaging. Since the radionuclidic impurities contribute significant effects on the patient's overall radiation dose, it is important to have a product with acceptable quality control (QC) parameters. Quality control (QC) parameters include tests for radiochemical, radionuclidic, and chemical purity for pharmaceutical concerns. The definition of radionuclidic purity is the ratio of the indicated radionuclide activity to the total radioactivity. In this study, the determination of radionuclidic impurities was done by sequential separation of target radionuclides ($^{89}$Sr, $^{90}$Sr and total alpha/beta emitters) followed by sensitive measurement using Liquid Scintillation Counting (LSC) system in the $^{99m}$Tc eluate from a $^{99}$Mo/$^{99m}$Tc generator. This sequential separation method was validated with standard solutions of the radionuclides used and the measurement uncertainty of the method was evaluated in detail. The uncertainty associated with the activity concentration has been calculated according to the appropriate rules of uncertainty propagation to give a combined standard uncertainty. The parameters contributing to the uncertainty budget include the uncertainties in the counting statistics, detector efficiency, misclassification (PSA), the mass of sample and activities of the standard radionuclide solutions. The major contributions come from the counting statistics as expected.

KEYWORDS: Radionuclidic impurity, Measurement uncertainty, $^{99m}$Tc, Liquid Scintillation Spectrometry

1. INTRODUCTION

Technetium-99m is a nuclear isomer of technetium-99. It is known as the most commonly used medical radioisotope because of its use in tens of millions of medical procedures annually. It is an almost ideal radionuclide for use in single photon emission computed tomography (SPECT) due to favorable decay characteristics. It decays by an isomeric transition (IT) with a half-life of 6.01 hours and emits $\gamma$-rays at 140 keV (89%). In a $^{99}$Mo/$^{99m}$Tc generator, $^{99m}$TcO$_4^-$ is formed by the decay of $^{99}$MoO$_4^{2-}$. The total concentration of technetium in the $^{99m}$Tc generator eluent is in the range of $10^{-7}$ to $10^{-6}$ M. In the eluate, $^{99}$Mo is the most important radionuclidic impurity and cause extraordinary radiation to the patient due to their relatively longer half-life (65.9 h) compared to $^{99m}$Tc (6 h). Therefore, it is required by the medical authorities that $^{99m}$Tc eluate from the $^{99m}$Tc-$^{99}$Mo generator has to contain a limited amount of radionuclides other than $^{99m}$Tc.
The radionuclidic purity is defined as the ratio between the activity of the base radionuclide and total activity of a radioactive compound, according to the European Pharmacopoeia [Ph. EU]. It is important in radiopharmacy since any radionuclidic impurities may increase the radiation dose received by the patient and may also degrade the quality of an imaging procedure performed. No radionuclide sample is 100% pure and all contain some contaminants arising from the production process or the decay of the primary radioisotope. In order to control the effects of these contaminants on the radiation dose received by the patient, limits must be set on the maximum levels of contamination allowed. These limits are defined in pharmacopeia monographs and vary depending upon the radionuclide concerned. According to European Pharmacy Monograph (Ph. EU), the major concerning radionuclides in the sodium pertechnetate injection ($^{99m}$Tc eluate) from a $^{99m}$Tc-$^{99}$Mo generator include $^{99}$Mo, $^{13}$I, $^{103}$Ru, $^{89}$Sr, $^{90}$Sr, alpha-emitting impurities, and other gamma-emitting impurities. Therefore, the determination of the radionuclidic purity at trace and ultra-trace levels must be done accurately and precisely before using these medical radioisotopes.

In addition, in any measurement, there is always a component of uncertainty due to systematic and random errors which should be fully considered. Uncertainty may be estimated from calculations of precision and bias. Precision is defined as the closeness of two or more measurements to each other. Bias may be defined as the estimate of a systematic measurement error (BIPM 2012). As a consequence, an analytical procedure for quality assessment often has a degree of uncertainty which can lead to false negative or false positive classification (Taylor et al. 2005; Boon and Ramsey, 2012). The estimation of uncertainty involves the identification, quantification, and summation of each potential source of uncertainty. The objectives of this work are therefore to determine the individual uncertainty components for the whole radioanalytical method, starting from the sample preparation up to the radioactivity measurement using the liquid scintillation spectrometry. For this purpose, the sequential separation steps of the method that contribute considerably to the combined measurement uncertainty were defined. The uncertainty associated with a certain parameter was used to determine its partial contribution to the combined total uncertainty. Finally, the uncertainty budget was expressed in a clear and simple way.

2. MATERIAL AND METHOD

2.1. Materials, standards and chemical reagents

Strong basic anion exchange resin AG1×4, Cl− form (50-100 mesh, Cat. No. A4-B500-C-Cl), Sr-resin in 2 mL plastic column (100-150 μm, Cat. No. AC-141-AL) and 2 mL plastic empty columns were purchased from Eichrom Technologies LLC, IL. 20 mL glass vials and liquid scintillation cocktail UltimaGoldLLT were purchased from Perkin Elmer Inc.( Massachusetts, USA). All chemicals used in the experiment were of analytical grade and prepared using deionized water. $^{85}$Sr, $^{90}$Sr, $^{90}$Y and $^{241}$Am standard solutions were purchased from National Institute of Standard and Technology (NIST, Gaithersburg, MD, USA).

2.2. Instruments

The gamma-ray spectrometric system has consisted of an n-type coaxial high purity germanium (HPGe) detector with 50% relative efficiency. Total alpha/beta, $^{85}$Sr, $^{90}$Sr, and $^{90}$Y isotopes activity measurements were performed using low background liquid scintillation
counter Wallac Quantulus 1200 TM (Perkin Elmer Life Science, Turku, Finland). The instrument is equipped with a pulse shape discrimination device (PSA) and an external standard of $^{152}$Eu which allows measuring external spectral quench parameter [SQP(E)].

2.3. Sequential separation procedure of the impurity radionuclides in the $^{99m}$Tc eluate

In this study, we used a sequential separation procedure for determining the levels of radionuclide impurities in Tc-$^{99m}$ eluates before use in patients (Xiaolin Hou, 2017). Since $^{99m}$Tc has a short half-life (6.02 h) compared to other impurity radionuclides, the primary eluate is allowed to decay for more than 7 days, so that the $^{99m}$Tc activity does not interfere with the measurement of trace amounts of radionuclidic impurities. Then the $^{99m}$Tc eluate of 5 mL was divided into two aliquots; one 2 mL aliquot was transferred to a 10 mL glass vial, which was stored in a lead pot. After 7-10 days decay, this aliquot was directly measured using $\gamma$-spectrometry for all gamma emitters including $^{99}$Mo, $^{131}$I and $^{103}$Ru. The removal of $^{99m}$Tc, $^{99}$Tc, and $^{99}$Mo is carried out by one step chemical separation using anion exchange chromatography.

2.4. Purification of strontium isotopes for measurement of $^{89}$Sr and $^{90}$Sr

If the activity of total beta is higher than $6 \times 10^{-6}$ % of the total activity with reference to the date and hour of eluting of $^{99m}$Tc, another aliquot from the anion exchange column is used to separate strontium for measurement of $^{90}$Sr and $^{89}$Sr. This aliquot was loaded to the Sr resin column preconditioned by 8M HNO$_3$. Strontium isotopes were eluted using 0.05 M HNO$_3$ solution and immediately measured by LSC, ($^{90}$Y in-growth is neglected). The same solution was re-measured after 6-10 days by LSC for Cherenkov counting of $^{90}$Y + $^{89}$Sr. Each sample was measured 30 min.

The chemical recovery of the separation procedure was calculated by using $^{241}$Am, $^{85}$Sr, $^{90}$Sr/$^{90}$Y and $^{90}$Y as tracers. A standard solution of these radionuclides was prepared by spiking the same amount of the individual radionuclide solution. The spiked solution was loaded to the AG1-X4 anion exchange column. The effluent and washes were collected in a vial and then divided into two aliquots. One 2 mL aliquot was directly measured using $\gamma$-spectrometry for $^{85}$Sr. For $^{241}$Am, $^{90}$Sr/$^{90}$Y and $^{90}$Y, the solution was mixed with UGLLT scintillation cocktail in a 20 mL glass vial, and measured by LSC. By comparison with the standard of each radionuclide, the chemical recovery of these radionuclides was calculated.

2.5. Determination of the total alpha/beta activity concentration

The optimum PSA levels were determined by calibrating with a set of pure $^{241}$Am and $^{90}$Sr/$^{90}$Y standard solutions. The vials were measured at PSA settings in a range from 50 to 180. The percent misclassification of $\alpha$-signals into the $\beta$-channel and vice versa ($\beta$-signals into the $\alpha$-channel) were calculated and plotted against the PSA values. The optimum PSA value was determined as 65. The following formula is used to calculate the total alpha and total beta activity concentrations in the samples.

$$\text{Activity (Bq/g)} = \frac{(N - N_{bkg})(cps)}{\varepsilon \times m \ (g)}$$ (1)
where, $N$ is the gross counting rate for the sample, in counts per second (cps), $N_{bkg}$ is the counting rate for the blank vials (cps), $\varepsilon$ is the counting efficiency and $m$ is the sample mass, in gram.

### 2.6. Determination of the activity concentration of strontium isotopes

$^{89}\text{Sr}$ can be quantified according to the following equation;

$$A_{89\text{Sr}} = \frac{(N - N_{bkg})}{\varepsilon_{89\text{Sr}}} \times R \times m$$

(2)

where $A_{89\text{Sr}}$ is the activity concentration of $^{89}\text{Sr}$, $N$ and $N_{bkg}$ are the total count rate (cps) and background count rate (cps) for the $^{89}\text{Sr}$ measurement, $R_{chem}$ is the chemical yield of the strontium separation, $\varepsilon_{89\text{Sr}}$ is the measurement efficiency for $^{89}\text{Sr}$ (cps/Bq). In order to calculate the activity concentration of $^{90}\text{Sr}$, the following equations were used:

$$A_{90\text{Y}} = \frac{(N - N_{bkg})}{\varepsilon_{90\text{Y}}}$$

(3)

where the parameters are the same as in Eq.(2) but for $^{90}\text{Y}$ instead of $^{89}\text{Sr}$. By calculating the activity of $^{90}\text{Y}$ at time $t_i$, i.e. the time allowed for ingrowth of $^{90}\text{Y}$ from $t_0$ (time of separation), one can determine the activity of $^{90}\text{Sr}$ according to:

$$A_{90\text{Sr}} = \frac{(A_{90\text{Y}}) \times (1 - e^{-\lambda_{90\text{Y}} \times \Delta t})}{(1 - e^{-\lambda_{90\text{Y}} \times \Delta t})} \times R_{chem}$$

(4)

where $A_{90\text{Sr}}$ is activity concentration of $^{90}\text{Sr}$ and $\lambda_{90\text{Y}}$ is the decay constants of $^{90}\text{Y}$ in s$^{-1}$, $\Delta t$ is the time interval between the separation of $^{89}\text{Sr}$ and the time at the end of counting in s$^{-1}$.

### 2.7. Calculation of the combined measurement uncertainty

The combined measurement uncertainties for total alpha/beta, $^{89}\text{Sr}$ and $^{90}\text{Sr}$ isotopes were calculated in accordance with the Guide to the Expression of Uncertainty in Measurement (GUM), ISO 1995 and the equations associated with calculating the activity from the total alpha/beta and Cherenkov measurements.

3. RESULTS AND DISCUSSION

#### 3.1. Uncertainty components in total alpha/beta activity determinations

The GUM (ISO, 1995) software was employed to determine the expanded uncertainty for each measurement using parameters already described to calculate activities (Equations 1 & 10) and main uncertainty sources identified for the radioanalytical method employed. Tables 1 to 3 present all uncertainty budgets obtained with statistical parameters and their contribution to the total uncertainty. The parameters contributing to the uncertainty budget of total alpha/beta activity concentration include the uncertainties in the counting statistics, detector counting efficiency, misclassification (PSA) and mass of the solutions.

Uncertainty in the net count rate is calculated by using the following equation.

$$Net\ count = N_{total\ counts} - N_{bkg}$$

(5)
\[ U_{\text{net count}} = \sqrt{N_{\text{total counts}} + N_{\text{bkg}}} \]  

(6)

In order to evaluate the variation on counting rates, 3 repeated measurements of one sample were done and the standard uncertainty was calculated and entered into the uncertainty budget. The same procedure was done for background uncertainty contribution. The uncertainty of the detector efficiency was estimated from a series of repeated observations by calculating the standard deviation of the mean (3 measurements for \(^{89}\)Sr and \(^{90}\)Sr efficiency and 10 measurements for total alpha/beta efficiency). The uncertainty associated with the PSA value was estimated using the misclassification equation given below;

\[
\% M_\beta = \frac{N_\beta \text{ in } \alpha \text{ MCA}}{N_\beta \text{ in } \beta \text{ MCA} + N_\beta \text{ in } \beta \text{ MCA}} \times 100
\]  

(7)

\[
\% M_\alpha = \frac{N_\alpha \text{ in } \beta \text{ MCA}}{N_\alpha \text{ in } \beta \text{ MCA} + N_\alpha \text{ in } \beta \text{ MCA}} \times 100
\]  

(8)

\[
\left[ \frac{U_{\text{misclassification}_{\beta}}}{\text{misclassification}_{\beta}} \right]^2 = \left[ \frac{\sqrt{\text{counts}_{12}}}{\text{counts}_{12}} \right]^2 + \left[ \frac{\sqrt{\text{counts}_{11} + \text{counts}_{12}}}{\text{counts}_{11} + \text{counts}_{12}} \right]^2
\]  

(9)

\[
\left[ \frac{U_{\text{misclassification}_{\alpha}}}{\text{misclassification}_{\alpha}} \right]^2 = \left[ \frac{\sqrt{\text{counts}_{11}}}{\text{counts}_{11}} \right]^2 + \left[ \frac{\sqrt{\text{counts}_{11} + \text{counts}_{12}}}{\text{counts}_{11} + \text{counts}_{12}} \right]^2
\]  

(10)

Table 1. Uncertainty budget and contribution of the major components for total alpha activity calculations in spike \(^{241}\)Am and \(^{90}\)Sr/\(^{90}\)Y solution.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Standard Uncertainty</th>
<th>Distribution</th>
<th>Uncertainty contribution, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net counts</td>
<td>2401</td>
<td>49.37</td>
<td>normal</td>
<td>2.06</td>
</tr>
<tr>
<td>Counting efficiency, (\varepsilon)</td>
<td>1</td>
<td>1</td>
<td>normal</td>
<td>1</td>
</tr>
<tr>
<td>Missclassification, PSA</td>
<td>0.0137</td>
<td>0.5326</td>
<td>normal</td>
<td>1.27</td>
</tr>
<tr>
<td>Chemical recovery, (R_{\text{chem}})</td>
<td>92.97</td>
<td>0.94</td>
<td>normal</td>
<td>1.59</td>
</tr>
<tr>
<td>Weighing, g</td>
<td>2.0248</td>
<td>0.00009</td>
<td>rectangular</td>
<td>0.0071</td>
</tr>
<tr>
<td>Activity, Bq.g(^{-1})</td>
<td>1.25</td>
<td></td>
<td></td>
<td>5.65</td>
</tr>
</tbody>
</table>

Table 2. Uncertainty budget and contribution of the major components for total beta activity calculations in spike \(^{85}\)Sr, \(^{241}\)Am and \(^{90}\)Sr/\(^{90}\)Y solution.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Standard Uncertainty</th>
<th>Distribution</th>
<th>Uncertainty contribution, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net counts</td>
<td>115843</td>
<td>341</td>
<td>normal</td>
<td>0.29</td>
</tr>
<tr>
<td>Counting efficiency, (\varepsilon)</td>
<td>0.95</td>
<td>0.75</td>
<td>normal</td>
<td>1</td>
</tr>
<tr>
<td>Missclassification, PSA</td>
<td>0.7814</td>
<td>0.1044</td>
<td>normal</td>
<td>1.27</td>
</tr>
<tr>
<td>Chemical recovery, (R_{\text{chem}})</td>
<td>81.26</td>
<td>0.97</td>
<td>normal</td>
<td>1.59</td>
</tr>
<tr>
<td>Weighing, g</td>
<td>2.0248</td>
<td>0.00009</td>
<td>rectangular</td>
<td>0.0071</td>
</tr>
<tr>
<td>Activity, Bq.g(^{-1})</td>
<td>65.58</td>
<td></td>
<td></td>
<td>2.57</td>
</tr>
</tbody>
</table>
Table 3. Uncertainty budget and contribution of the major components for Sr-89 and Y-90 activity calculations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Uncert. contribution, % ((^{89}\text{Sr}))</th>
<th>Uncert. contribution, % ((^{90}\text{Y}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Counting statistics (incl. weight)</td>
<td>13.43</td>
<td>10.14</td>
</tr>
<tr>
<td>Counting efficiency, (\varepsilon)</td>
<td>1.27</td>
<td>3.99</td>
</tr>
<tr>
<td>Chemical recovery, (R_{\text{chem}})</td>
<td>4.51</td>
<td>4.51</td>
</tr>
<tr>
<td>Weighing, g</td>
<td>0.0021</td>
<td>0.0021</td>
</tr>
<tr>
<td>Combined standard uncertainty (quadratic sum), (\sqrt{\sum ((u(x)/x)^2)})</td>
<td>(k = 1)</td>
<td>(k = 1)</td>
</tr>
</tbody>
</table>

For total alpha and beta emitters, the uncertainty mainly comes from the statistic error of LSC counting in the sample, blank and standards, the spillover of beta counts to the alpha window and alpha counts in the beta window, counting efficiency, recovery of alpha and beta emitters in separation procedure and sample weight. For \(^{89}\text{Sr}\) and \(^{90}\text{Sr}\), statistic error of LSC counting, counting efficiency calibration, recovery in the chemical separation steps (anion exchange and Sr-column chromatography) and sample weight are the main components.

The sequential separation method was used for the analysis of \(^{99m}\text{Tc}\) eluate obtained from the 4 batches of the \(^{99}\text{Mo}/^{99m}\text{Tc}\) generator for radionuclidic impurity determination and the results were given in Table 4. The measured analytical impurities are lower than the limitation of European pharmacopeia [Ph. EU]. Uncertainties presented in the analytical results are expanded uncertainties using a coverage factor \(k=2\), which was estimated considering all possible contributions. Since the content of \(^{89}\text{Sr}\), \(^{90}\text{Sr}\), and total \(\alpha\)-emitters are very low in all samples, the analytical uncertainties are relatively high. This is mainly because of the counting uncertainty for samples and blanks.

Table 4. Analytical results of impurity radionuclides in the \(^{99m}\text{Tc}\) eluate from some batches of \(^{99}\text{Mo}/^{99m}\text{Tc}\) generators for quality control

<table>
<thead>
<tr>
<th>Sample code</th>
<th>(^{99m}\text{Tc}) GBq/mL</th>
<th>Gross Beta</th>
<th>Ratio, %</th>
<th>Gross Alpha</th>
<th>(^{89}\text{Sr})</th>
<th>(^{90}\text{Sr})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tc_1</td>
<td>1.012</td>
<td>528.28 (13.58)</td>
<td>5.22×10(^{-6})</td>
<td>10.15 (0.57)</td>
<td>1.00×10(^{-6})</td>
<td>0.484 (0.069)</td>
</tr>
<tr>
<td>Tc_2</td>
<td>1.028</td>
<td>272.17 (6.99)</td>
<td>2.65×10(^{-6})</td>
<td>5.32 (0.39)</td>
<td>5.17×10(^{-7})</td>
<td>0.387 (0.055)</td>
</tr>
<tr>
<td>Tc_3</td>
<td>1.022</td>
<td>56.34 (1.45)</td>
<td>5.51×10(^{-6})</td>
<td>2.77 (0.16)</td>
<td>2.71×10(^{-7})</td>
<td>1.815 (0.258)</td>
</tr>
<tr>
<td>Tc_4</td>
<td>1.022</td>
<td>103.41 (2.66)</td>
<td>1.03×10(^{-6})</td>
<td>5.59 (0.32)</td>
<td>5.57×10(^{-7})</td>
<td>3.548 (0.505)</td>
</tr>
</tbody>
</table>

**Ratio:** The radionuclidic purity is defined as the ratio between the activity of the base radionuclide and total activity of a radioactive compound.

### 4. CONCLUSION

Radionuclide impurities can have a significant impact on the patient's overall radiation dose and may affect image quality. In this study, the determination of radionuclidic impurities was done by sequential separation of \(^{89}\text{Sr}\), \(^{90}\text{Sr}\) and total alpha/beta emitters in \(^{99m}\text{Tc}\) eluate obtained from the \(^{99}\text{Mo}/^{99m}\text{Tc}\) generator. The total alpha/beta and \(^{89}\text{Sr}\), \(^{90}\text{Sr}\) activities were measured using LSC by employing \(\alpha/\beta\) discrimination function and Cherenkov radiation, respectively. The measurement uncertainty components were evaluated in detail and it is seen that the major sources of uncertainty were identified as uncertainties associated with the counting rates as expected.
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ADSORPTION STUDIES of Ce(III) onto THE MULTI-WALLED CARBON NANOTUBES by USING RESPONSE SURFACE METHODOLOGY

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ABSTRACT

The scope of this study is to investigate the adsorption effectiveness of modified multi-walled carbon nanotubes (MWCNTs) on cerium(III) ions. Two different modifications (HNO₃ and H₃PO₄) were applied. Parameters effecting the adsorption “contact time, pH, initial Ce(III) concentration and temperature” were examined and optimum conditions determined by using Central Composite Design (CCD) technique.

KEYWORDS: Cerium, Adsorption, Carbon Nanotubes, Response Surface Methodology

1. INTRODUCTION

The demand for rare earth elements is increasing day by day as it has widespread use in the latest technology areas such as engineering, information storage, communications, automobiles, metallurgy and nuclear energy. This increase in demand causes more release of rare earth elements to the environment. Rare earth elements can lead to significant negative impacts on health, such as genetic poisoning in bone marrow cells and DNA damage. Cerium element is one of the cheapest and abundant rare earth elements which has widespread use in industry in polishing powder and ceramics, catalyst support, pyrophoric alloys and metallurgical fields. When cerium reaches to living organisms, it usually accumulates in the liver, lungs and kidneys of the human body (Hao et al., 2019; Kütahyali et al., 2012). Therefore, many methods are applied to remove cerium from liquid wastes (ion exchange, solvent extraction, adsorption, chemical precipitation, membrane filtration and electrochemical process). Adsorption is the most widely used and cost-effective process which eliminates the disadvantages such as; secondary waste generation and low selectivity of the other separation methods (Farghali et al., 2013).

Carbon nanotubes are one of the most effective adsorbents which used in adsorption process. Carbon nanotubes are relatively new materials that were first discovered by lijima in 1991 and have superior properties in graphite structure. Since their discovery, they have attracted great attention with their unique chemical, physical and mechanical properties. They have a large surface area, porous and layered structure. The use of carbon nanotubes as sorbent materials in the environmental area gives promising results in the management and treatment of liquid wastes (Belloni et al., 2009).

In this study, cerium adsorption on modified MWCNTs was investigated. MWCNTs were modified with nitric acid and phosphoric acid to increase the uptake capacity by formation of functional groups on the surface. Optimum conditions were defined by central
composite design technique as a function of contact time, temperature, pH and initial cerium concentration.

2. MATERIAL AND METHODS

All chemicals used in the study were obtained from Merck. MWCNTs were obtained from Sigma-Aldrich, with the following characteristics: 10 nm outer and 4.5 nm internal diameter; 4 µm in length and purity about 70-80%.

MWCNTs were modified with nitric acid (HNO₃) and phosphoric acid (H₃PO₄). In nitric acid modification (NA-MWCNTs), MWCNTs added to the 8 mol/L HNO₃ solution and stirred for 16 hours at room temperature. Then the solution was filtered through a 0.10 µm membrane filter and modified adsorbent washed with distilled water until neutral pH. Drying was carried out at 100°C (Belloni et al., 2009). To modify with phosphoric acid, MWCNTs were soaked in 80% of H₃PO₄ solution and stirred during 4 hours at room temperature. After filtration was performed, modified MWCNTs with H₃PO₄ (PA-MWCNTs) washed until neutral pH and dried at 100°C (Yakout and Sharaf El-Deen 2016). GFL 1086 temperature controlled thermostat shaker was used at 130 rpm for the adsorption studies. Experiments were carried out according to experimental design matrix which defined by using central composite design technique. The following equation was used to determine the amount of cerium adsorbed on modified MWCNTs:

\[ q = (C_0 - C_e) \times \frac{V}{m} \] (mg.g⁻¹)

where \( q \) is the uptake capacity (mg.g⁻¹), \( C_0 \) and \( C_e \) are the initial and equilibrium concentrations in solution (mg.L⁻¹), \( V \) is the solution volume (mL) and \( m \) is the mass of adsorbent (g).

3. RESULTS AND DISCUSSION

Effect of time on cerium adsorption with multi-walled carbon nanotubes were carried out as preliminary experiments and optimum conditions which is defined, used in the CCD matrix experimental set. Figure 1 shows the effect of time on adsorption. According to results, optimum time was identified as 15 minutes for NA-MWCNTs and 30 minutes for PA-MWCNTs.
The initial pH of the solution is an important factor for adsorption studies due to the influencing of the surface charge and metal species. Figure 2 shows binary interaction of investigated parameters on cerium adsorption with modified multi-walled carbon nanotubes. According to figure 2, uptake capacity increases as pH and initial Ce(III) concentration increase. This can be related to the higher availability of cerium ions for sorption which causes the higher driving force for making utilization of active sides effective (Abuh et al., 2013). The optimum conditions which defined by investigating pH, initial cerium concentration, temperature and contact time were listed in Table 1.
Table 1. Optimum conditions for adsorption of Ce(III) onto the modified MWCNTs

<table>
<thead>
<tr>
<th>Parameters</th>
<th>NA-MWCNTs</th>
<th>PA-MWCNTs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial Ce(III) concentration (mg.L⁻¹)</td>
<td>167</td>
<td>167</td>
</tr>
<tr>
<td>pH</td>
<td>6</td>
<td>5</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>25</td>
<td>29</td>
</tr>
<tr>
<td>Contact time (min.)</td>
<td>15</td>
<td>30</td>
</tr>
<tr>
<td>Uptake Capacity (mg.g⁻¹)</td>
<td>380.21</td>
<td>392.55</td>
</tr>
</tbody>
</table>

4. CONCLUSION

In this study, adsorption of cerium with modified MWCNTs has been investigated as a function of time, pH, initial cerium concentration and temperature by using batch experiment technique. Central composite design model was used to analyze the adsorption model and optimization of significant factors. Our results confirm that, MWCTNs have potential for applications which used in nuclear waste management. The adsorption properties of carbon nanotubes make this material promising for the treatment of waste from nuclear plants.

5. REFERENCES


ASSESSMENT OF THE RADIOLOGICAL IMPACT OF COAL-FIRED POWER PLANTS TO TERRESTRIAL BIOTA USING ERICA TOOL

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ABSTRACT

In this article, the radiological effects of coal-fired power plants (CFPP) on terrestrial and marine environments are evaluated and the risk of radiation exposure to non-human biota is evaluated. Radiological evaluations were made considering the reported radionuclide activity concentrations. The radiation dose rates that reference organisms may be exposed to were calculated using the ERICA Tool. The results showed that lichen and bryophytes can receive the highest doses in terrestrial ecosystem. In the aquatic ecosystem, insect larvae, zooplankton and molluscs were found to be exposed to the highest radiation dose.

KEYWORDS: non-human biota, coal mine, radiological impact, dose risk assessment

1. INTRODUCTION

Biota can be exposed to both external and internal ionizing radiation from radionuclides in various ways. In recent years, the determination of the effects of radiation, especially on non-human biota, has gained importance (UNSCEAR, 2008; Brown et al., 2008; ICRP, 2009; USDoe, 2002; IAEA, 2014).

To evaluate the ecological risk of ionizing radiation in terrestrial, freshwater and marine biota, the ERICA assessment tool developed within the scope of a project (Beresford et al. 2007) has been used in different studies (Konstantinova et al., 2015; Vandenhove et al., 2015).

The coal industry is one of the activities that causes the increase in natural radioactivity in environmental media. The coal can contain uranium, thorium and ⁴⁰K from natural radioactive series. According to UNSCEAR (1982) and UNSCEAR (2008) reports, the average activity concentrations of ²³⁸U and ²³²Th in the coal are 20 Bq kg⁻¹ (in the range of 5-300 Bq kg⁻¹).

The aim of this study was to evaluate the exposed dose rates of terrestrial and aquatic biota with ERICA Tool by using the natural radioactivity values obtained from TAEK Technical Reports on the coal, soil and sediment samples taken from the coal-fired power plants (CFPPs) in different provinces in Turkey.

2. MATERIAL AND METHOD

In this study, the external and internal dose rates evaluations were made by using the activity concentrations compiled from TAEK Atlas (2014) and TAEK Report (2011) for the coal, soil and sediment samples taken from the coal-fired power plants (CFPPs) in Bursa, Çanakkale, Kahramanmaraş, Kütahya, Manisa, Muğla, Sivas and Zonguldak.
The ERICA Tool version 1.2, which is based on a layered approach in three separate steps, was used to assess radiological risk from coal for non-human biota (Brown et al., 2016). Some reference terrestrial and marine organisms are defined to the ERICA Tool to estimate the exposed radiation dose rate for these organisms.

3. RESULTS AND DISCUSSION

Mean radioactivity concentrations in soils were used as data to assess the impact on terrestrial and aquatic ecosystems. Cesium-137 activity concentrations in the sediment are not reported in the TAEK Coal Technical Report. Therefore, concentrations of $^{226}$Ra, $^{232}$Th and $^{137}$Cs were used only in the soils and $^{226}$Ra and $^{232}$Th concentrations in the sediments.

The highest concentration of $^{226}$Ra in the soil was observed in Çanakkale. The highest concentration of $^{232}$Th in the soil was observed in Kütahya and the highest concentration of $^{137}$Cs in the soil was observed in Sivas. In the sediments, the highest concentrations of $^{226}$Ra and $^{232}$Th were observed in Yatağan CFPP. The highest concentration of $^{226}$Ra in the coals was observed in Kangal CFPP and the highest $^{232}$Th in Yatağan CFPP.

According to the average activity concentrations in soil, the highest exposed doses to the terrestrial reference organisms were observed in lichen and bryophytes. The activity concentrations of the aquatic reference organisms in the marine ecosystem due to concentrations of $^{226}$Ra and $^{232}$Th were found to be the highest in Yatağan CFPP. The highest activity concentrations were observed in insect larvae and mollusc both bivalve and gastropod.

When the total absorbed dose rates for the terrestrial reference organisms resulting from $^{226}$Ra, $^{232}$Th and $^{137}$Cs were examined, the highest total dose rates among the organisms were obtained for lichen and bryophytes, grass & herbs and shrubs. The total dose rate resulting from $^{226}$Ra was found to be maximum in lichen and bryophytes in Çanakkale and minimum in tree in Muğla in the range of $3.41 \times 10^{-2}$ - $3.63 \mu$Gy h$^{-1}$. The total dose rate caused by $^{232}$Th was calculated at the maximum value in the lichens and bryophytes in Kütahya in the range of $6.17 \times 10^{-5}$ - $4.00 \times 10^{-4} \mu$Gy h$^{-1}$ and the minimum value in large mammals in Zonguldak.

The effect of $^{226}$Ra concentrations on total doses in CFPPs was more dominant in the sediment. The maximum dose rate value of $^{226}$Ra in insect larvae was determined in Yatağan CFPP in the range of $1.01 \times 10^{-4}$ – $81.4 \mu$Gy h$^{-1}$. The minimum value was obtained in Soma CFPP for mammals.

4. CONCLUSION

For terrestrial organisms, exposure to maximum doses ($3.625 \mu$Gy h$^{-1}$) resulting from the activity concentrations of $^{226}$Ra in the soils are predominate in lichens and bryophytes.

For aquatic organisms, exposure to the highest doses with a dose rate of about $82 \mu$Gy h$^{-1}$ in the Yatağan CFPP site is thought to be the case for insect larvae. This is followed by zooplankton and molluscs.
5. REFERENCES


NATURAL AND ANTHROPOGENIC RADIOACTIVITY OF PLANT AND ANIMAL ORGANISMS IN A MOUNTAIN ECOSYSTEM

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ABSTRACT

The study presents a radioecological investigation on monitor plants and monitor small rodents species from two different altitudes in Rila Mountain (South-West Bulgaria): 1500 m a.s.l. and 2925 m a.s.l.

Plant species – Poa alpine, Sesleria coerulans and Epilobium parviflorum, and small rodents species – yellow necked mouse (Apodemus flavicollis), bank vole (Myodes (Clethrionomys) glareolus) and snow vole (Chionomys nivalis), were used to estimate the environmental state using radiometric methods.

The radiometric measurements were carried out by using a low-background beta counter (LAS 3A low-level activity system with 30 % efficiency on ⁴⁰K).

The comparison with the data of the total beta-activity revealed the existence of differences between the bioaccumulation in the organism of the different monitor species from the two areas. The results showed that the total beta-activity in plants ranged from 394 Bq kg⁻¹ to 654 Bq kg⁻¹. In Apodemus flavicollis from Moussala Peak (2925 m a.s.l.), the values varied from 144 Bq kg⁻¹ up to 309 Bq kg⁻¹; and from 168 Bq kg⁻¹ up to 491 Bq kg⁻¹ for the lower altitude (1500 m a.s.l.). For Myodes glareolus these values reached 1081 Bq kg⁻¹. Measurements from the bodies of the Chionomys nivalis from Moussala peak were between 203 Bq kg⁻¹ and 623 Bq kg⁻¹.

The results obtained can be considering as normal. The data demonstrate significantly lower values in comparison with the same measurements twenty years ago.

KEYWORDS: Plants, Small rodents, Total beta-activity, Rila Mountain
1. INTRODUCTION

Mountains are particularly important objects of studies on the impact of various natural and anthropogenic factors on the natural ecosystems. Organisms develop in constantly changing conditions of radiation of natural and anthropogenic origin (Jovtchev et al., 2019). Its effect on living organisms is yet to be thoroughly studied.

Living organisms are exposed to external irradiation by mainly cosmic, electromagnetic and neutron radiation, and to internal irradiation caused by radioisotopes incorporated during respiration and with water and food. The radioisotopes in soil and water are uptaken by plant roots and are thus transported to all tissues and organs.

We report the results of high-mountain biomonitoring in Rila Mountain performed at Moussala Basic Environmental Observatory (BEO) located on Moussala Peak 42°10’45"N, 23°35’07"E, 2925 m a.s.l.), the highest peak of Rila Mountain and the Balkan Peninsula.

To assess the possible environmental contamination using radiometric methods, species of plants and small mammals were used as biomonitors, namely, the plant species Poa alpina, Sesleria coerulans and Epilobium parviflorum and the small rodent’s species yellow necked mouse (Apodemus flavicollis), bank vole (Myodes (Clethrionomys) glareolus) and snow vole (Chionomys nivalis).

We followed the total beta-activity as an indicator of beta-emitting isotopes in different environmental samples, which then could be used for radiological assessment of different sources of pollution. The primary sources of beta-activity are the naturally occurring radioisotopes ($^{226}$Ra, $^{228}$Ra, $^{228}$Th) from the natural decay series of uranium, thorium and actinium; the primordial isotope, $^{40}$K; and any residual anthropogenic fallout from, e.g., weapons testing and the Chernobyl reactor accident in 1986, as $^{137,134}$Cs and $^{90}$Sr (Metcheva et al., 2008, Damianova et al., 2014, Almayahi et al., 2012, Tonev et al., 2016).

Changes in the radiation background have become important in recent years, especially after the Chernobyl (IAEA 2001; Yordanova et al. 2007) and Fukushima accidents (Itoh et al. 2014).

The aim of the present study was to investigate the natural and anthropogenic radioactivity in plants and small rodents that are specific for ecosystems at different altitudes in Rila Mountain (1500 m a.s.l. and 2925 m a.s.l.).

The measurements are part of a project for Complex Monitoring of Rila Mountain performed by the Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences.

2. MATERIAL AND METHOD

2.1. Sampling

The sampling was conducted in the summer of 2017. In habitats at the two quoted altitudes in Rila Mountain, the plant species collected were as follows: Poa alpina L. (Poaceae), Sesleria coerulans Friv. (Poaceae), Epilobium angustifolium L. (Onagraceae); and the following small rodent species were captured: yellow-necked mouse (Apodemus flavicollis) (n=13) and bank vole (Myodes (Clethrionomys) glareolus) (n=22) from Skakavtsite, and the snow vole (Chionomys nivalis) (n=12) and the yellow-necked mouse (Apodemus flavicollis) (n=12) from Moussala Peak.

The specimens of the different monitor species of small rodents were caught using live baits and snap traps. The traps were checked every 24 h. The captured live mice were transported to the laboratory.
2.2. Analytical procedure

The samples were treated according to a standard procedure: mineralization at 400 °C for 4 hours and measurements of material (13.26 mg cm$^{-2}$) by means of a LAS 3A low-level activity system (30% efficiency on $^{40}$K and background 1 cpm$^{-1}$) (Iovtchev et al., 1996). The plant samples for laboratory analyses were dried at 110 °C to a constant weight and then ashed. All collected animals were dissected to remove the intestinal tract with contents, as well as most internal organs and tissues – livers, kidneys, spleens, and bones. The samples were oven-dried at 60 °C for approximately three days to obtain a reference mass and analyzed for total beta-activity (Iovtchev et al., 1997).

3. RESULTS AND DISCUSSION

Table 1 presents in Bq kg$^{-1}$ the mean values of the total beta-activity of different monitor species of plants and small rodents as ashed material:

Table 1. Total beta-activity in different monitor species of plants and small rodents from the two investigated areas in Rila Mountain.

<table>
<thead>
<tr>
<th>Moussala</th>
<th>$A$ (mean ± SD) [Bq kg$^{-1}$]</th>
<th>Skakavtsite</th>
<th>$A$ (mean ± SD) [Bq kg$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Poa alpina</td>
<td>413 ± 11</td>
<td>Epilobium parviflorum</td>
<td>654 ± 12</td>
</tr>
<tr>
<td>Sesleria coerulans</td>
<td>394 ± 12</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Apodemus flavicollis</td>
<td>230.3 ± 7.2</td>
<td>Apodemus flavicollis</td>
<td>366.3 ± 8.1</td>
</tr>
<tr>
<td>Chionomys nivalis</td>
<td>382.0 ± 8.3</td>
<td>Myodes glareolus</td>
<td>524.2 ± 5.3</td>
</tr>
</tbody>
</table>

The plants grown at 1500 m a.s.l. showed a total beta-activity of 654 ± 12 Bq kg$^{-1}$. The plant material sampled on Moussala Peak at 2925 m a.s.l. included representatives of P. alpina and S. coerulans, where we found a relatively low beta-activity in Poaceae (413 ± 11 Bq kg$^{-1}$ in P. alpina and 394 ± 12 Bq kg$^{-1}$ in S. coerulans).

Differences were observed in the total beta-activity of mice and voles in both investigated areas, which can be explained by the difference in their food specialization. Mice, such as Ap. flavicollis, are omnivorous, while voles are mainly herbivorous species. The values of the total beta-activity in Skakavtsite area are higher in comparison with the data for Moussala peak, since Beli Iskar Skakavtsite area is near a closed uranium mine and the area is rich in granites.

For the first time, the total beta-activity of monitor plants and animal species from Rila Mountain was measured in the 1995-1997 period. The results obtained then showed a significantly higher beta-activity of plant species (~ 5000 – 13000 Bq kg$^{-1}$ for grasses) than those presented here (~ 400 – 1300 Bq kg$^{-1}$). The same applies to the animal species (Iovtchev et al. 1995, 1996). Figure 1 illustrates a comparison of the data:
The main contributor to the formation of the total beta-activity is $^{137}\text{Cs}$. In Bulgaria, as in all countries in South-Eastern Europe, the deposition of $^{137}\text{Cs}$ originates mainly from the fallout from the Chernobyl accident (Tonev et al, 2016). Recent studies (2015) have indicated that the amount of $^{137}\text{Cs}$ in the surface soil layer of (0 – 5 cm) is comparable to that before the accident in Chernobyl power plant. This decrease is mainly due to the natural decay of $^{137}\text{Cs}$ and to a very small degree to migration processes in depth of the soil (Yordanova et al., 2016).

As the results show, the levels of total beta-activity in the species studied plants and small rodents from Rila Mountain were relatively low and can be taken as a normal for the regions investigated.

4. CONCLUSION

In the framework of the project for complex monitoring of Rila Mountain, plants and small rodents were investigated from two different altitudes and areas. Data were obtained on the total beta-activity of the plant species *Poa alpina*, *Sesleria coerulans* and *Epilobium*, and of small rodents the yellow-necked wood mouse (*Apodemus flavicollis*), bank vole (*Clethrionomys glareolus*) and snow vole (*Chionomys nivalis*). The total beta-activity values observed are lower than these from the 20 years ago.

These results can be considered as normal and thus used for further monitoring and tracing the accumulation on different levels of the trophic chains in the ecosystems.

ACKNOWLEDGEMENTS

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Fig.1. Comparison between the values of total beta-activity in the body of monitor species of small rodents from 1997 and 2017
5. REFERENCES


INTERACTION OF CESIUM AND STRONTIUM IONS WITH ENGINEERING BARRIER MATERIAL AND THEIR DOSE RISK ASSESSMENT

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ABSTRACT

This study deals with performance of removal of cesium (Cs⁺) and strontium (Sr²⁺) ions from synthetic aqueous solution using amino pyridine sulfone amid resin as a barrier material. The effects of adsorbate concentration, temperature and contact time on the engineering barrier material efficiencies of Cs⁺ and Sr²⁺ were investigated and evaluated. Furthermore, the suitability of the barrier system to be designed a simulated waste storage area was examined radiologically. The radiological dose risk assessment of cesium and strontium that will diffuse into the barrier system after an accident was performed by using ERICA Tool software. In the program, the results were examined based on some reference organisms.

KEYWORDS: nuclear waste, barrier material, sorption, interaction, cesium, strontium, biota

1. INTRODUCTION

Nuclear power plants, one of the important sources of energy production, produce high levels of nuclear waste and these wastes need to be stored. The storage of waste can be ensured by being buried in specially selected deep underground depots. Such nuclear waste landfills usually rely on a multi-barrier system to isolate waste from the biosphere. Multiple barrier systems include natural rock or minerals and engineering barrier systems (Andryushchenko et.al., 2017). Using these barrier systems, long-life radionuclides in waste can be stopped before they reach the ecosystem (Fuks et.al., 2019). The presence of radionuclides such as cesium-137 (t½=30.1 years) and strontium-90 (t½=28.5 years) with long life and high fission efficiency is the main source of radiotoxicity in high-level wastes (Cheremisina et.al., 2017).

Aim of the study are to investigate the usage possibility of an engineering barrier system to removal of cesium and strontium in the event of a leak in waste storage, to clarify the mechanism of interaction of cesium and strontium wastes in the engineering barrier system using different models for the design of adsorption systems, to calculate thermodynamic parameters, such as enthalpy of adsorption (ΔH), free energy change (ΔG), and entropy change (ΔS) from adsorption experiment data in the different temperature range to determine the spontaneity of interaction process, and to evaluate the effects of chronic exposure to

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radiation of fresh water, marine and terrestrial biota in case of a leakage from waste storage areas.

2. MATERIAL AND METHOD

High purity aluminium oxide, styrene, divinylbenzene, benzoyl peroxide, toluene, chlorosulfonic acid and amino pyridine was used to synthesis amino pyridine sulfonamide resin (APSR). The convenience of the obtained material as a barrier material for simulated nuclear waste deposition area was tested with cesium nitrate and strontium nitrate solutions and adsorption experiments. Double distilled water was used throughout the study.

Adsorption experiments were carried in a thermostated shaker water bath by using batch method. Adsorption process was investigated for different concentration, temperature, and contact time. After the adsorption, adsorption solutions were analysed for cesium and strontium by atomic absorption spectrometer. The percentage removal of Cs\(^+\) and Sr\(^{2+}\) and the adsorption capacity \(q_e\) of the APSR were calculated using initial and equilibrium concentration of adsorbate.

The adsorption process on the surface of the prepared adsorbent was investigated with Langmuir, Freundlich, Dubinin Radushkevich and Temkin isotherm models.

Kinetics of sorption describing the solute uptake rate, which in turn governs the contact time, were investigated by applying pseudo first order, pseudo second order and Elovich rate models to experimental data.

The ERICA Tool software system (Beresford et al. 2007; Larsson, 2008), that has a structure based upon the tiered ERICA Integrated Approach to assessing the radiological risk to terrestrial, freshwater and marine biota, was used to calculate total exposure dose to non-human biota.

3. RESULTS AND DISCUSSION

Different adsorption isotherm models such as Langmuir, Freundlich, Dubinin-Radushkevich and Temkin were applied the adsorption data. Evaluating the adsorption data, it was found that the data fits to the Dubinin-Radushkevich adsorption isotherm \(R^2=0.99\) which indicates that the adsorption is on single type of uniform pores.

The effect of temperature on the interaction of cesium and strontium ions with the prepared adsorbent was investigated. Adsorption reactions were found as typically exothermic for both ions. Therefore, in accordance with Le Chatelier’s principle, the magnitude of adsorption should increases with decreasing in temperature. This indicates that adsorption is physical adsorption.

Thermodynamic parameters such as enthalpy, entropy and Gibbs free energy chance were estimated from the slopes and intercepts of the plots of ln \(K_d\) vs. I/T. Gibbs free energy was calculated by using Gibbs equation. It was found that adsorption enthalpy is exothermic for both Sr and Cs ions. The positive value of entropy change indicates the increased randomness at the solid-solution interface during the adsorption of strontium and cesium on the barrier.
system. The negative values for the Gibbs free energy change show that the adsorption process is thermodynamically feasible.

Three kinetic models were applied to adsorption kinetic data in order to investigate the behaviour of interaction process of Cs and Sr onto barrier material. These models are the pseudo-first-order, the pseudo-second-order and the intraparticle diffusion models. Based on the obtained correlation coefficients ($R^2$), the experimental data conformed better to the pseudo-second-order equation, evidencing chemical sorption as rate-limiting step of adsorption mechanism. The pseudo-second-order model is based on the assumption that the rate-limiting step may be chemisorption involving valence forces through the sharing or exchange of electrons between adsorbent and adsorbate.

Calculations of total exposure dose to non-human biota were performed primarily with the ERICA tool software. The results showed that totally external and internal doses for fresh water biota will be high in insect larvae, crustacean and vascular plant from Cs and Sr activity without using the prepared barrier material. After using barrier material, the total dose per organism is significantly decreased. The totally external and internal doses for marine biota were calculated high in polychaete worm, macroalgae and corals from Cs and Sr activity without the barrier material. The total dose per organism is significantly decreased using the barrier material. The results showed that totally external and internal doses for terrestrial biota will be high in deer, reptile, rat and lichen\&bryophytes from Cs and Sr activities without using the barrier material. It was observed that the barrier material has caused to significantly fall the total dose per organism.

4. CONCLUSION

The amino pyridine sulfonamide resin was synthesized and applied as adsorbent for removal of Cs$^+$ and Sr$^{2+}$ ions from aqueous solution. The percentage removal of Sr$^{2+}$ ions onto APSR are better than Cs$^+$ ions. According to these results, adsorbate concentration, temperature and contact time on the engineering barrier material are important parameters effecting Cs$^+$ and Sr$^{2+}$ removal.

The ERICA integrated approach indicated impacts of radioactivity on the environment. Further research is in progress to determine optimum removal conditions.

ACKNOWLEDGEMENTS

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5. REFERENCES


VERIFICATION OF GAMMA SPECTROMETRY ANALYTICAL RESULTS USING SIMULATED AND EXPERIMENTALLY DERIVED EFFICIENCY CURVES

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ABSTRACT

A correct efficiency curve is a crucial element to accurately determine the activity contents in different environmental matrices using gamma spectrometry techniques. Different critical parameters (such as sample’s geometry, type of radionuclide, density, and chemical composition of the samples) are affecting the accuracy of the calibration efficiency curve. The most recommended method to calculate such a curve is the experimental method using similar matrix of a certified reference material. The experimental approach is usually expensive and time-consuming, primarily when a wide range of samples with different matrices and radionuclides are analyzed. An alternative method is to generate the efficiency calibration curve using either the semi-empirical or a numerical calculation, which is based on photon interaction with the materials and the detector-sample geometry arrangements. This work presents an approach to experimentally verify the efficiency curves generated by LabSOCS (Laboratory Sourceless Object Calibration Software) of various environmental sample matrices. Different IAEA PT samples were analyzed using the efficiency curves generated by the two methods. The analytical results were compared, and the necessary correction factors for cascade summing were applied. A comparison between the efficiency curves generated experimentally and by LabSOCS has been carried out. The results showed that the efficiency curves generated by LabSOCS can be used to determine the concentration of different radionuclides with less than 10% bias from the target values for water samples and less than 20% for soil samples. It was found that the elemental composition of the sample and the parameters of the counting geometry should be accurately and precisely defined in the software to obtain accurate and precise results, in particular when analyzing low energy gamma emitters of certain radionuclides (241Am, 234Th, and 210Pb). The advantages of using the LabSOCS were to apply the cascade summing corrections, in addition, to accurately determine the activity of low energy gamma emitters in different samples’ matrices.

KEYWORDS: Simulation efficiency, radioactivity measurement, cascade summing correction

1. INTRODUCTION

Gamma spectrometry is one of the practical techniques which allows both qualitative and quantitative analysis natural and artificial radionuclides in various types of environmental samples (Bell et al., 2012; Miguel et al., 2002; Walling, 2002). It has several advantages of the other techniques of measuring environmental radioactivity such as the simplicity of sample preparation that can replace radiochemical separation procedures with nondestructive measurements. Other advantages are related to the accuracy, reliability, and sensitivity. However, the sensitivity and accuracy can be affected by the efficiency curve used to quantifying the radionuclides presenting in the sample. the efficiency curve does not depend only on the system characterization but also on other factors such as the sample density and chemical (elemental) composition, the sample-detector geometrical arrangement, and the photon energy of the radionuclides (Aba and Ismaeel, 2013; Kaminski et al., 2014; Misiak et al., 2011). Furthermore, the efficiency curve is very sensitive to the coincidence summing phenomenon of the gamma lines which are in cascade (Mauring and Drefvelin, 2012).
Several methods of efficiency calibration for gamma spectrometry are in use in radioactivity measurement laboratories. The most recommended method is the experimental method, wherein an ideal situation the calibration source is identical to the measured sample. However, in routine work of such laboratories, the preparation of the calibration sources are approximating as closely as possible to the geometry (container, filling the height of the sample and sample-detector arrangement, and sample’s matrix). Although this method is recommended, it is expensive and time-consuming especially when measuring a wide range of samples with different materials. Another method is to prepare a smaller number of calibration standard solutions in different containers with different heights, and if the samples are different with respect to matrix and density, a self-attenuation correction factor can be applied in addition to the cascade summing correction. However, several software codes are available to transfer efficiencies measured from one geometry to another (Kaminski et al., 2014).

The recent method is the mathematical efficiency calibrations that are based on the theoretical calculation of the photons interaction probabilities with the detector. This requires full information of the physical characteristics of the detector and the counting geometry. In this method, there is no need for reference material preparation; the efficiency can be directly generated by using a mathematical calculation based on Monte-Carlo simulation of the photon interaction with the materials. LabSOCS is an example of such software that is being used in KISR radioactivity measurements laboratory. It is a patented computer program performing mathematical efficiency calibrations of HPGe detectors, without any use of radioactive sources (Bronson, 2003). Such software has many advantages; it saves time and money spent on purchasing and measuring certified reference materials of different geometries. It also can generate efficiency curves for different and unique types of geometry. The main objective of this study was to experimentally test the validity of utilizing the efficiency curves generated, and the cascade summing correction performed by LabSOCS.

2. MATERIAL AND METHOD

Samples of IAEA inter-comparison tests (IAEA-PT-2014, IAEA-PT-2015) and samples of Proficiency Test of ERA Company (A Waters Company) in 2016 were used in this study as test samples with known activity concentrations. Detailed information and radionuclides contained in these samples are listed in Table 1.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Detailed</th>
<th>Studied Radionuclides</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample-1</td>
<td>IAEA-PT-2014-400g water</td>
<td>$^{241}$Am, $^{226}$Ra, $^{134}$Cs, and $^{137}$Cs</td>
</tr>
<tr>
<td>Sample-2</td>
<td>IAEA-PT-2014-400g water</td>
<td>$^{134}$Cs, $^{137}$Cs, and $^{210}$Pb</td>
</tr>
<tr>
<td>Sample-3</td>
<td>IAEA-PT-2014-400g water</td>
<td>$^{241}$Am, $^{226}$Ra, and $^{152}$Eu</td>
</tr>
<tr>
<td>Sample-4</td>
<td>IAEA-PT-2015-200g soil</td>
<td>$^{40}$K, $^{238}$U, $^{232}$Th, and $^{137}$Cs</td>
</tr>
<tr>
<td>Sample-5</td>
<td>ERA-PT-200g water</td>
<td>$^{241}$Am, $^{238}$U, $^{134}$Cs, $^{137}$Cs, $^{60}$Co, and $^{65}$Zn</td>
</tr>
<tr>
<td>Sample-6</td>
<td>ERA-PT-300g soil</td>
<td>$^{228}$Ac, $^{241}$Am, $^{212}$Bi, $^{214}$Bi, $^{134}$Cs, $^{137}$Cs, $^{60}$Co, $^{40}$K, $^{212}$Pb, $^{214}$Pb, $^{234}$Th, $^{238}$U, $^{65}$Zn</td>
</tr>
</tbody>
</table>

Two different gamma spectrometry systems (UltraLow Background Spectrometry system [ULB] and N-type gamma spectrometry system) were used to measure these samples. Both spectrometry systems are Canberra Company products; equipped with (HPGe) detectors. And the Gamma analytical software developed by Canberra (Genie-2000) was used for gamma spectra analyses.
Two methods were used to calculate the efficiency curve using in determining the activity concentrations for each radionuclide in the test samples. First, experimentally efficiency curves were calculated using different certified reference materials of different geometries. Table 2 shows the detailed information of the certified reference samples used.

<table>
<thead>
<tr>
<th>Eff-Cali.file</th>
<th>Source</th>
<th>Radionuclides</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exp-1-water</td>
<td>NPL</td>
<td>$^{241}\text{Am}$, $^{57}\text{Co}$, $^{137}\text{Cs}$, $^{54}\text{Mn}$, $^{88}\text{Y}$, $^{65}\text{Zn}$, and $^{60}\text{Co}$</td>
</tr>
<tr>
<td>Exp-2-water</td>
<td>$^{226}\text{Ra}$ CRM</td>
<td>$^{226}\text{Ra}$</td>
</tr>
<tr>
<td>Exp-3-water</td>
<td>QC IAEAPT 2006</td>
<td>$^{241}\text{Am}$, $^{60}\text{Co}$, $^{134}\text{Cs}$, $^{137}\text{Cs}$, and $^{210}\text{Pb}$</td>
</tr>
<tr>
<td>Exp-4-water</td>
<td>QC IAEAPT 2014</td>
<td>$^{241}\text{Am}$, $^{226}\text{Ra}$, $^{134}\text{Cs}$, and $^{137}\text{Cs}$</td>
</tr>
<tr>
<td>Exp-5-water</td>
<td>QC IAEAPT 2015</td>
<td>$^{152}\text{Eu}$</td>
</tr>
<tr>
<td>Exp-6-water</td>
<td>ERA QC</td>
<td>$^{241}\text{Am}$, $^{238}\text{U}$, $^{137}\text{Cs}$, $^{60}\text{Co}$, and $^{65}\text{Zn}$.</td>
</tr>
<tr>
<td>Exp-1-soil</td>
<td>IAEA-447</td>
<td>$^{241}\text{Am}$, $^{228}\text{Ac}$, $^{137}\text{Cs}$, $^{212}\text{Pb}$, $^{210}\text{Pb}$, $^{226}\text{Ra}$, $^{40}\text{K}$, and $^{238}\text{U}$.</td>
</tr>
<tr>
<td>Exp-2-soil</td>
<td>ERA QC</td>
<td>$^{228}\text{Ac}$, $^{241}\text{Am}$, $^{212}\text{Bi}$, $^{214}\text{Bi}$, $^{134}\text{Cs}$, $^{137}\text{Cs}$, $^{60}\text{Co}$, $^{40}\text{K}$, $^{212}\text{Pb}$, $^{214}\text{Pb}$, $^{234}\text{Th}$, $^{238}\text{U}$, and $^{65}\text{Zn}$</td>
</tr>
</tbody>
</table>

Second, four efficiency curves (for water samples of 200 and 400 ml and for soil samples of 80 and 200 g) were generated by using LabSOCS software for both spectrometry systems. However, the characterization file including the required information of the detector and the geometry dimension and type of material should be defined for each test sample in order to run the software. The different activity concentration of each test samples was compared with the target value of the samples given by the PT organizations (IAEA, ERA Company). The relative bias between the calculated activity value and the target value (B) has been calculated using equation 1:

$$B = \frac{V - T}{T} \times 100$$

(1)

Where V is the reported value and T is the target value.

To accept the results of activity concentration the B values should be less than $kP$, where k is the coverage factor for 95% confidential level and equal to 2.56, and $P$ is the relative combined uncertainty and can be calculated by the following equation (IAEA criteria please add a reference):

$$P = \sqrt{\left(\frac{U_T}{T}\right)^2 + \left(\frac{U_V}{V}\right)^2} \times 100$$

(2)

3. RESULTS AND DISCUSSION

Verification of LabSOCS Gamma Efficiency Simulation-free CSC. The activity concentrations of radionuclides of free cascade summing of gamma energies ($^{40}\text{K}$, $^{137}\text{Cs}$ $^{210}\text{Pb}$, $^{212}\text{Pb}$, $^{226}\text{Ra}$, $^{228}\text{Ac}$, $^{234}\text{Th}$, $^{65}\text{Zn}$, and $^{241}\text{Am}$) in the test samples were calculated using different experimental efficiency curves, and simulation efficiency curve generated by LabSOCS software for different geometries. The relative bias values for each efficiency calibration curve used to measure activity concentration of some radionuclide are shown in figure 1. Results show that the activity concentration calculated by using experimental efficiency curves that contains similar radionuclides in test samples or have closed gamma energy lines were accepted. Nevertheless, missing data points in the efficiency cross over curve will affect the results of gamma energy lines less than 200 KeV, such as $^{226}\text{Ra}$ (186 KeV) concentration measured by EXP-3 efficiency curve. IAEA-447 certified reference material (moss sample) was used to generate experimental the efficiency curve. Since the (chemical compassion) is different between the moss and soil samples, an attenuation correction factor $C_{\text{att}}$ was applied.
for the low energy lines (46 keV ($^{210}$Pb) and 63 keV ($^{234}$Th). Equation 3 was used to calculate the correction factors, which were found to be 1.145 and 1.16 for 46 and 63 keV, respectively. On the other hand, all results of activity concentration for the measured radionuclides calculated based on LabSOCS simulation curve were accepted; taking into account the chemical compassion.

$$C_{at} = S \cdot \frac{\ln \left( \frac{\tilde{C}}{S} \right)}{1 - \left( \frac{S}{\tilde{C}} \right)}$$

(3)

Where $C_{at}$: Corrected values of measured count per second of the sample, $S$: measured CPS of the sample, $\tilde{C}$: the attenuated intensity of the source, and $\tilde{S}$: ($\tilde{C}$) Through the sample

**Verification of LabSOCS Gamma Efficiency Simulation- with CSC.** The activity concentrations of radionuclides with multiple gamma lines that required cascade summing correction ($^{60}$Co, $^{134}$Cs, $^{152}$Eu, $^{214}$Bi, and $^{214}$Pb) were calculated using different experimental efficiency and simulation curves. $^{134}$Cs concentration results generated by the experimental efficiency curves were not accepted that because $^{134}$Cs was missing in the Exp1 and Exp2 efficiency curve. Therefore, the simulated efficiency curves were used and the results were accepted. The similar discussion can be adopted for the $^{60}$Co and $^{152}$Eu activity concentration calculation.

**4. CONCLUSION**

As a conclusion the procedure of LabSOCS simulation software can be applied for determining multi radionuclides activity of different environmental matrices; including radionuclides with cascade summing. The critical parameters that have to be considered for
accurate and precise determination are: the chemical composition of the sample matrix and the sample to geometry arrangements, in addition to a precise detector configuration.

REFERENCES


SUBMITTING THE PAPER
The full paper has to be submitted by email to insinume2019@gmail.com.
DISTURBANCE OF HYDROLOGICAL REGIME AS A FACTOR OF PARTICULATE Cs-137 MIGRATION IN THE UPA RIVERTITLE

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ABSTRACT

Lateral migration of $^{137}$Cs via temporary and constant watercourses is the main mechanism for spatial transformation of initially deposited contaminants on Chernobyl affected territories. In Central Russia, highly contaminated areas occur on densely populated territories where watercourses are exposed to major human impacts. As far as the highest concentration of $^{137}$Cs in soils and deposits can be attributed to the Chernobyl fallout, the floodplain strata turn out to be very important of landscape record. Depth increment study of floodplain deposits provides information about $^{137}$Cs concentration in suspended sediments during flood events in the post-Chernobyl period. The analysis of floodplain morphology and topography can reveal modern trends of river valley development. This information can be used for interpretation of obtained radiocesium data. Moreover, accumulation of sediments on the floodplains in contaminated areas illustrates the process of natural attenuation, which should be considered in remediation strategy development. Studies conducted in 2009-2018 in the Upa River basin (right-hand tributary of the Oka River) have made it possible to identify trends in particulate $^{137}$Cs lateral migration in different reaches of the river. The most part of sediment-associated $^{137}$Cs delivered from the cultivated slopes of the Upa River upper reaches have been trapped in Schekino reservoir. As the result, incision is observed downstream and accumulation of sediment-associated $^{137}$Cs on the floodplains is decreasing dramatically. In the middle reaches, the sedimentation rates on floodplain returned to normal. The low reaches of the Upa River are characterized by significant incision caused by intensive excavation of sands from the channel of the Oka River since the last decades of 20th century.

KEYWORDS: $^{137}$Cs, Chernobyl contamination, floodplains, sediments, lateral migration

1. INTRODUCTION

Water erosion and transport of eroded material with temporary and constant water streams is a basic mechanism of particulate Cs-137 lateral migration. Human impact accelerates this process by intensive cultivation of top soils on the interfluvies and mid-slopes, where bigger part of fallout Cs-137 is deposited. In lowlands of temperate climatic zone the major part of eroded sediments are usually in the lower part of cultivated slopes or in dry valleys, gullies or artificial sinks (Fridman et al., 1997; Golosov, 2002). Rivers is the major way for distant migration of particulate $^{137}$Cs after Chernobyl fallout, as well for all contaminants associated with sediments (Vetrov et al., 1990). Disturbance of hydrologic regime by human activity is
argued as an important factor of sediment redistribution and migration of particulate $^{137}$Cs. In the Upa River basin human activities which have serious impact on hydrological regime can be subdivided in two types. The first one is construction of artificial reservoirs, which decrease inclination on water surface upstream and trap major part of transported sediments. As a result, downstream from the dam water stream has lack of sediments and intensive erosion of river bed occurs. The biggest artificial reservoir in the Upa River basin is located near Shekino power plant.

The second type is represented by river sand mining of construction materials in the Oka River, the receiving water course the Upa River. Intensive excavation of sands and gravels in the middle of 20th century on the Oka River resulted into drop of water level and intensive erosion downstream by the quarries and in down reaches of its tributaries, including the Upa River (Berkovich et al., 2012).

2. MATERIAL AND METHODS

The observed basin is situated in forest-step landscape zone and can be characterized as a typical catchment for Central Russia with high share of cultivated areas. After Chernobyl accident in 1986 the territory of the basin was contaminated with $^{137}$Cs (“Plavsk hot spot”), basically the upper reaches of the Upa River and its left-bank tributaries. A series of points was chosen on the floodplain of the Upa River, where depth increment sampling procedure was undertaken in 2009-2018. Two additional sampling sites was appointed on the Oka River floodplain upstream and downstream from the mouth of the Upa River. Samples were dried, weighed, grinded and gamma-spectrometry analysis measured with spectrometer ORTEC GEM-C5060P4-B with HPGe detector (relative efficiency 20%).

3. RESULTS AND DISCUSSION

As it was discussed previously reservoirs are acting as a sediment traps, decreasing turbidity of water and as a result rates of overbank accumulation. Concentration of $^{137}$Cs in sediments is very suitable record, which illustrated dynamics of sediments transportation and migration of particulate radionuclides. In particular $^{137}$Cs is basically transported with fine particles, especially after intensive erosion on arable slopes during snowmelt in spring. In opposite, the bulkier particles are deposited on floodplains during high-water periods. Under undisturbed conditions (PIR-1 sampling site) the vertical distribution of $^{137}$Cs in floodplain strata can be characterized by presence of layer with maximal concentration of radionuclides, which can be attributed to 1986 (Fig 1A). Inventories of $^{137}$Cs above the can be assumed as accumulated after 1986. According to direct observations and concentration of $^{137}$Cs after 1986 drops dramatically (Vakulovskii et al., 1996). This tendency is well illustrated by distribution of radionuclides in sediments accumulated over post-Chernobyl period, including sections on floodplains. The situation downstream by the reservoir looks different (MEZ-1 sampling site). The $^{137}$Cs is distributed more chaotic in the upper part of the strata and no particular layer can be reliably attributed to 1986 (Fig 1B). Moreover it looks like concentration of $^{137}$Cs almost didn’t drop for the post-Chernobyl time but even raised. This can be explained by following. The reservoir may not catch the finest suspended particles, which actually can adsorb significant amounts of radionuclides. If flux of suspended material from reservoir was stable in time the decreasing of $^{137}$Cs concentration may not occur.
Figure 1. Vertical distribution of $^{137}$Cs in floodplain sediments from sites upstream (A) and downstream (B) from Sheckino reservoir

In down reaches of the Upa River accumulation and radionuclides on floodplains is very variable process. As it seen from sections Up-2 and Up-1 tributaries can raise inventories of $^{137}$Cs almost in ten times at adjacent part of receiving river. Moreover, rates of accumulation within explored area also can differ in several times. For the major part of explored sections rates turned out to be surprisingly high. Even maximal concentration of $^{137}$Cs corresponded to 1986 was detected at list in 3 of 8 sections (Fig 2).

Figure 2. $^{137}$Cs distribution on floodplains in the downreaches of the Upa River.

Topography of river valley’s bottom is critical for overbank accumulation. On shallow floodplains big amounts of sediments can be deposited during flooding, but this volumes are redistributed over relatively large area. Hence, mean rates of accumulation can be rather moderate. In opposite case, when after intensive incision, floodplain occupies narrow part of the valleys bottom, rates can growth in times, because accumulation occurs in rather compact zone.
4. CONCLUSION

Through example of the Upa River, it was observed that anthropogenic impact results into different changes in sediment yield and intensity of radionuclides’ migration. Accumulation of particulate $^{137}\text{Cs}$ in artificial sinks may cause emergence of new risks in case of their demolition. However, human impact may have positive results linked to intensive accumulation of comparably clean sediment over contaminated strata and as a result natural attenuation.

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5. REFERENCES

REFERENCES


TECHNOGENIC RADIONUCLIDES DISTRIBUTION IN GRAIN-SIZE FRACTIONS OF SOIL IN THE AREA OF EXCAVATION NUCLEAR TEST (“ATOMIC LAKE”)

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ABSTRACT

The paper presents research findings of artificial radionuclides (137Cs, 90Sr, 241Am, 239 240Pu) distribution in grain-size fractions of soil in the area of the “Atomic Lake” artificial water reservoir created due to an excavation explosion at Semipalatinsk Test Site. It was revealed that fission products (137Cs, 90Sr) and elements of a nuclear charge material (241Am, 239+240Pu) have different pattern of distribution in grain-size fractions of the soil. 137Cs and 90Sr are predominately accumulated in the finest soil fraction (<1 µm), whereas 241Am and 239+240Pu radionuclides are mostly accumulated in coarser fractions (1000-500 µm, 40-8 µm). It was observed that the pattern of radionuclides distribution in grain-size fractions of soil can reflect features of radioactive contamination of the area generated by the excavation nuclear explosion.

KEYWORDS: soil, grain-size fraction, technogenic radionuclide, excavation nuclear explosion, Semipalatinsk Test Site

1. INTRODUCTION

Nuclear excavation (cratering) explosion is the type of an underground one during which movement and ejection of earth material from the nuclear device detonation point to the surrounding landscape take place in a relatively short time (Koranda et. al., 1970; Logachev, 2001). The first excavation explosion (“Chagan” explosion, 15.01.1965) at Semipalatinsk Test Site was aimed at obtaining information about possibilities of using nuclear explosions for creating water reservoirs in arid areas. The “Chagan” explosion was conducted in the hole 1004 in the “Balapan” testing area. The water storage reservoir called as the “Atomic lake” was created as the result of the explosion (Akchurin, 2007; Logachev, 1997, 2001; Shkolnik, 2003). Radioactive contamination of soil is mainly spread to the area with the radius of 4-5 km adjacent to the explosion epicenter (Aidarkhanov et. al., 2017).

2. MATERIAL AND METHOD

Object of research. As the objects of research the radioactive fallout plume that spread in the north-western direction (from the outside crater area) were chosen.

Research methodology. The research methodology consisted of soil sampling, separation of samples to grain-size fractions and determination of concentration of radionuclides in soil samples and fractions.

Sampling. Total 13 topsoil samples (5 cm depth) were collected along the radioactive fallout plume. Sampling points were located in the direct line with increasing distance from the...
explosion epicenter. The distance between sampling points was from 60 to 400 m. The most distant sampling point was located 2700 m away from the explosion epicenter.

Fractions separation. The techniques of «wet» sieving and sedimentation in a suspension were used for separation of soil into grain-size fractions. Therefore, by using the «wet» sieving method the 1000-500; 500-250; 250-100; 100-63 and 63-40 µm fractions were separated. The fractions with grain size of less than 40 µm were subsequently separated by applying the sedimentation method. In general, each sample was divided to 9 grain-size fractions.

Radionuclide analyses. The activity concentration of artificial radionuclides was determined in soil samples and grain-size fractions. Activity concentration of $^{137}$Cs and $^{241}$Am were determined using Canberra GX-2020 gamma-spectrometer with semiconductor detector. $^{90}$Sr activity concentration was determined by direct measuring via «Progress-BG» beta-spectrometer (Russia). However, if activity concentration of the radionuclide was below the detection limit (100 Bq/kg), the $^{90}$Sr activity concentration was determined after application of radiochemical analysis. The radiochemical procedure for extraction and isolation of $^{90}$Sr and $^{239+240}$Pu is formed, briefly, by several stages: leaching out of radionuclides from the soil matrix by complete chemical decomposition, removal of interfering radionuclides, preparation of counting samples and determination of the corresponding activity concentrations. The $^{90}$Sr activity concentration was determined from the activity concentration of Y-90 by using a liquid scintillation beta-spectrometer “TRI CARB 3100 TR”. The activity concentrations of $^{239+240}$Pu in the sources were determined by using an «Alpha – Analyst» alpha-spectrometer («CANBERRA», USA), equipped with solid-state PIPS detectors.

3. RESULTS AND DISCUSSION

The microaggregate composition of soils. The mass percentage (ω, %) of each grain-size fraction of soil related to the total weight of the dried soil aliquot taken to the grain-size analysis was determined. The results of the grain-size analysis of soil samples collected at the research object are shown in the Table 1. Mean value of the mass percentage for each fraction is detailed.

<table>
<thead>
<tr>
<th>Object</th>
<th>Fraction, µm</th>
<th>1000-500</th>
<th>500-250</th>
<th>250-100</th>
<th>100-63</th>
<th>63-40</th>
<th>40-8</th>
<th>8-5</th>
<th>5-1</th>
<th>&lt;1</th>
</tr>
</thead>
<tbody>
<tr>
<td>plume</td>
<td></td>
<td>18,9</td>
<td>20,9</td>
<td>20,2</td>
<td>5,1</td>
<td>4,1</td>
<td>6,3</td>
<td>10,3</td>
<td>5,0</td>
<td></td>
</tr>
</tbody>
</table>

Results show that a microaggregate composition of soils predominantly consists of coarse fractions from 1000 µm to 100 µm. Microaggregates of finer than 100 µm include relatively higher content of 40-8 µm fraction. However, in rare cases it can be observed that soil enriched with 5-1 µm fraction. The content of silt fraction (<1 µm) is non-uniform. In some cases the mass percentage of silt fraction can reach 8-9%, however, from some samples the silt fraction failed to be separated.

Concentration of artificial radionuclides in soil. Results show that the fission products ($^{137}$Cs, $^{90}$Sr) make a major contribution to radioactive contamination of soil on the radioactive fallout plume. Despite the high contamination level, the activity concentration of transuranic elements ($^{239+240}$Pu, $^{241}$Am) is significantly lower than fission products. Specific activity of
$^{137}$Cs and $^{90}$Sr vary from n×$10^1$ to n×$10^4$ Bq/kg, while for $^{241}$Am and $^{239+240}$Pu this range lies within n×$10^1$ to n×$10^3$ Bq/kg. The spatial distribution of radionuclides along the plume is non-uniform. The highest concentration of radionuclides is observed at a distance of 300-500 m away from the explosion epicenter.

**Distribution of radionuclides in the grain-size fractions.** The so-called enrichment factor (Ef) has been used as a no-dimensional parameter to evaluate the $^{137}$Cs, $^{241}$Am, $^{239+240}$Pu and $^{90}$Sr distribution in the different soil grain-size fractions. The enrichment factor (Ef) was calculated for each soil fraction and each radionuclide of interest as the ratio between the activity concentration of the radionuclide in the fraction ($A_{fr}^{sp}$) and the activity concentration determined for the same radionuclide in the bulk soil aliquot ($A_{sp}^{s}$) corresponding to the fraction considered (Equation 1).

$$\text{Ef} = \frac{A_{fr}^{sp}}{A_{sp}^{s}}$$

This parameter serves as an index of enrichment or depletion of grain-size fractions relative to the average concentration of radionuclides in soil. Ef > 1 indicates that fraction is enriched by radionuclides, Ef < 1 indicates that the fraction is depleted by radionuclides. The enrichment factors values for the different radionuclides and the different fractions of the soils are compiled in the Table 2. Mean and range values for each fraction and each radionuclide of interest are detailed.

<table>
<thead>
<tr>
<th>Fraction, µm</th>
<th>$^{137}$Cs range</th>
<th>$^{137}$Cs mean</th>
<th>$^{90}$Sr range</th>
<th>$^{90}$Sr mean</th>
<th>$^{241}$Am range</th>
<th>$^{241}$Am mean</th>
<th>$^{239+240}$Pu range</th>
<th>$^{239+240}$Pu mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000-500</td>
<td>0.4 - 1.7</td>
<td>0.7</td>
<td>0.5 - 1.4</td>
<td>1.0</td>
<td>0.6 – 4.8</td>
<td>1.7</td>
<td>0.6 – 3.0</td>
<td>1.5</td>
</tr>
<tr>
<td>500-250</td>
<td>0.5 – 1.0</td>
<td>0.8</td>
<td>0.6 – 1.2</td>
<td>0.9</td>
<td>0.5 – 2.0</td>
<td>1.2</td>
<td>0.3 – 3.1</td>
<td>1.2</td>
</tr>
<tr>
<td>250-100</td>
<td>0.6 – 1.3</td>
<td>1.0</td>
<td>0.7 – 1.0</td>
<td>0.9</td>
<td>0.5 – 1.5</td>
<td>1.0</td>
<td>0.3 – 1.5</td>
<td>1.0</td>
</tr>
<tr>
<td>100-63</td>
<td>0.2 – 1.3</td>
<td>1.0</td>
<td>0.7 – 1.0</td>
<td>0.9</td>
<td>0.4 – 1.7</td>
<td>1.0</td>
<td>0.4 – 1.2</td>
<td>0.8</td>
</tr>
<tr>
<td>63-40</td>
<td>0.6 – 1.8</td>
<td>1.1</td>
<td>0.6 – 1.2</td>
<td>1.0</td>
<td>0.5 – 2.9</td>
<td>0.8</td>
<td>0.2 – 7.3</td>
<td>1.2</td>
</tr>
<tr>
<td>40-8</td>
<td>0.7 – 2.5</td>
<td>1.4</td>
<td>0.7 – 1.8</td>
<td>1.1</td>
<td>0.3 – 3.4</td>
<td>1.2</td>
<td>0.3 – 4.4</td>
<td>1.3</td>
</tr>
<tr>
<td>8-5</td>
<td>0.9 – 2.2</td>
<td>1.5</td>
<td>0.1 – 2.4</td>
<td>1.0</td>
<td>0.1 – 1.7</td>
<td>0.6</td>
<td>0.1 – 2.8</td>
<td>0.8</td>
</tr>
<tr>
<td>5-1</td>
<td>1.0 – 2.8</td>
<td>1.8</td>
<td>0.9 – 2.9</td>
<td>1.5</td>
<td>0.1 – 2.0</td>
<td>0.4</td>
<td>0.1 – 1.3</td>
<td>0.4</td>
</tr>
<tr>
<td>&lt;1</td>
<td>1.2 – 4.4</td>
<td>2.5</td>
<td>1.1 – 4.4</td>
<td>2.2</td>
<td>0.1 – 2.4</td>
<td>0.7</td>
<td>0.1 – 4.0</td>
<td>0.7</td>
</tr>
</tbody>
</table>

According to the results $^{137}$Cs and $^{90}$Sr distribution tends to the radionuclides concentration increase as the fraction size decrease. Coarse soil fractions of larger than 250 µm are mostly depleted by $^{137}$Cs and $^{90}$Sr. The highest value of the enrichment factor for $^{137}$Cs and $^{90}$Sr has revealed in the <1 µm fraction.

In general, the pattern of $^{241}$Am and $^{239+240}$Pu distribution in grain-size soil fractions tendency is opposite to the pattern of $^{137}$Cs and $^{90}$Sr distribution. Mainly the coarse soil fractions the size of larger than 250 µm are more enriched by $^{241}$Am and $^{239+240}$Pu. However, along the plume the pattern of $^{241}$Am and $^{239+240}$Pu distribution in grain-size fraction of soils is dissimilar. Thus, with distance from the explosion epicenter, 40-8 µm fraction becomes the most enriched by $^{241}$Am and $^{239+240}$Pu. According to Logachev (2001) with distance from ground zero, the median size of radioactive fallout particles that deposited on the surrounding area and resulted in radioactive contamination of the environment sharply decreased.
4. CONCLUSION

Technogenic radionuclides distribution in grain-size soil fractions in radioactive fallout plume formed around the venue of the "Chagan" excavation explosion at the Semipalatinsk Test Site has been studied.

Fission products ($^{137}$Cs and $^{90}$Sr) and elements of charge material ($^{241}$Am and $^{239+240}$Pu) distribution pattern were ascertained being different. However, both were found to be predominantly accumulated in the definite soil fractions, obviously due to radioactive fallout particles incorporated therein. Thus, the plume has both coarse (500-1000 $\mu$m) and fine (40-8 $\mu$m) soil fractions enriched with $^{241}$Am and $^{239+240}$Pu radionuclides, and the finest (<1 $\mu$m) fraction enriched with $^{137}$Cs and $^{90}$Sr.

Distribution of $^{241}$Am and $^{239+240}$Pu in the grain-size soil fractions have been found being significantly changed in the pattern along the plume. It can be concluded that radionuclides distribution in grain-size soil fractions in conjunction with the information on the environment contamination formation, can reflect the features of radioactive contamination generated by the excavation explosion.

5. REFERENCES


DEVELOPMENT OF ACTIVITY RATIOS RESEARCH METHOD FOR THE $^{241}$Pu AND $^{241}$Am IN THE SOIL OF THE MAIN STS TEST SITES

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ABSTRACT

Studies were conducted on the basis of which a radiochemical method was developed for the determination of $^{241}$Pu with an instrumental ending on a TRICARB liquid scintillation beta spectrometer. The developed method was tested on various STS test sites. The paper presents the obtained data on activity levels of $^{241}$Pu and concentration ratios of $^{241}$Pu / $^{241}$Am.

KEYWORDS: Semipalatinsk Test Site (STS), plutonium, americium, radiochemical.

INTRODUCTION

The radioecological situation at the Semipalatinsk test site (STS) is well studied and, it would seem, over time should only be improved due to the disintegration of the dose-forming radionuclides. However, there is a fact of an increase in the activity of one of the main dose-forming radionuclides - $^{241}$Am. $^{241}$Am is a decay product of $^{241}$Pu, the content of which in weapons-grade Pu is an order of magnitude greater than the activity of $^{239}$Pu, which is "habitually" determined in environmental objects due to the relative simplicity and popularity of the radiochemical method. Possessing a relatively short half-life (14 years), $^{241}$Pu quickly decays into $^{241}$Am ($T_{1/2} = 432$ years). In about 60 years on the STS, the activity of $^{241}$Am will reach its maximum value, which, given the half-life, will not change in the near future. Over the same time period, the activity of such elements as $^{90}$Sr and $^{137}$Cs will decrease by fourfold. Thus, the contribution of $^{241}$Am to the radiological situation of the STS over time will only increase. To qualitatively predict changes in the radiological situation over time, it is necessary to know the distribution of $^{241}$Pu in the STS at the moment.

Defining a $^{241}$Pu task is quite complicated. During the entire existence of the landfill, no more than a few dozen definitions were conducted, and these data are not known to us. Not all leading radiochemical laboratories in the world have $^{241}$Pu determination techniques. For a detailed study of the distribution of $^{241}$Pu in the landfill, taking into account its area, it is necessary to analyze several tens of thousands of samples. Taking into account the cost of analysis and delivery of samples to a foreign laboratory, such studies are inexpediently expensive [P. Thakur et al., 2017]. A significantly cheaper option for estimating the concentration of $^{241}$Pu is to calculate it by the concentration of $^{241}$Am, which is a decay product of $^{241}$Pu. However, this is possible if the concentration ratio of $^{241}$Pu / $^{241}$Am is known and is constant at least within the local STS sites.

The aim of this work was to study the relationship of the activity of $^{241}$Pu and $^{241}$Am in the soil of the main STS test sites.
MATERIAL AND METHOD

To successfully apply the liquid scintillation analysis method [M. Abuzwida et al., 1987], it is necessary to purify Pu isotopes from other beta-emitting radionuclides present in the sample. This procedure is well known, but given Pu’s polyvalence, its implementation is impossible without the use of a tracer. Also, when using the method of liquid scintillation, it is necessary to take into account the amendment to the "quenching" of the sample, and this is possible with the use of a standard solution of $^{241}$Pu.

In our case, it is proposed to use those isotopes of plutonium that are already in the sample as tracer. The method is based on carrying out a liquid scintillation beta-spectrometric analysis of a sample with a known activity of alpha-decaying plutonium isotopes - $^{239} + ^{240}$Pu and $^{238}$Pu. The activity of $^{239} + ^{240}$Pu and $^{238}$Pu is determined by the standard alpha spectrometric method. Such an analysis scheme implies thorough homogenization of the sample and is applicable only to samples with a total of $^{239} + ^{240}$Pu and $^{238}$Pu not less than 0.2 Bq per sample. To clean a beta-spectrometric sample from possible tritium, after standard radiochemical cleaning procedures, the sample is evaporated to dryness and tightly closed to prevent contact with the surrounding air [Ryan T.P et al., 1993].

For research, surface soil samples (0-5 cm layer) were taken from the most typical test sites of the former STS:
- test sites in the galleries of the "Degelen" mountain range; the location of the excavation explosion or "Atomic Lake" site; testing site for "Combat Radioactive Substances" (CRS); "Experimental field" and the territory in its zone of influence.

RESULTS AND DISCUSSION

Galleries of the "Degelen" mountain range.

The range of the obtained ratios of $^{241}$Pu / $^{241}$Am for each of two separately taken galleries lies in a rather narrow interval. Table 1 presents the results of determining $^{241}$Pu and $^{241}$Am and their ratio in soil samples taken at the "Degelen" site.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{241}$Am, Bq/kg</th>
<th>$^{241}$Pu, Bq/kg</th>
<th>$^{241}$Pu/$^{241}$Am</th>
<th>$^{241}$Pu/$^{241}$Am</th>
</tr>
</thead>
<tbody>
<tr>
<td>Шт-177/1</td>
<td>940 ± 65</td>
<td>17570 ± 3515</td>
<td>18.7 ± 4.7</td>
<td>18.8±4.7</td>
</tr>
<tr>
<td>Шт-177/2</td>
<td>810 ± 60</td>
<td>15335 ± 3070</td>
<td>18.9 ± 4.7</td>
<td></td>
</tr>
<tr>
<td>Шт-143/1</td>
<td>410 ± 36</td>
<td>2066 ± 413</td>
<td>5.0 ± 1.3</td>
<td>3.8±1.0</td>
</tr>
<tr>
<td>Шт-143/2</td>
<td>746 ± 65</td>
<td>1920 ± 385</td>
<td>2.6 ± 0.6</td>
<td></td>
</tr>
</tbody>
</table>

The date of the test in the 177 gallery was March 30, 1983, the theoretically calculated ratio of $^{241}$Pu / $^{241}$Am today is equal to 16.2 with an average of 18.8 ± 4.7.

The date of testing in the gallery 143 October 30, 1976, the theoretically calculated ratio of $^{241}$Pu / $^{241}$Am today is 10.5, with an average of 3.8 ± 1.0.

This difference between the analysis results and the theoretical calculation for the 143 adit is fully explained by the following series of facts:
- The purpose of the test in the gallery 177 - "The creation or improvement of nuclear weapons."
- The purpose of the test in the gallery 143 - "Study of the damaging factors of nuclear weapons and their impact on military and civilian objects."
From here it is possible to assume with a high degree of confidence that the newly created "fresh" charge was used for testing in the gallery 177, and the serial "tested", manufactured for a relatively long time, was used for the testing in the gallery 143. A reverse theoretical calculation showed that the possible creation time of the charge is 1958-1959. i.e. charge lay in storage for about 18 years. This fully satisfies the storage time of a nuclear charge, according to some data it is about 30 years.

The "Atomic lake" site.
Table 2 presents the results of determining $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratio in soil samples taken at the "Atomic lake" site.

Table 2. Results of determination of $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratios in soil samples taken on the site "Atomic lake".

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{241}\text{Am}$, Bq/kg</th>
<th>$^{241}\text{Pu}$, Bq/kg</th>
<th>$^{241}\text{Pu}/^{241}\text{Am}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>AO-01</td>
<td>344 ± 28</td>
<td>1430 ± 286</td>
<td>4.2 ± 1.0</td>
</tr>
<tr>
<td>AO-02</td>
<td>1090 ± 83</td>
<td>3094 ± 619</td>
<td>2.8 ± 0.7</td>
</tr>
<tr>
<td>AO-03</td>
<td>533 ± 48</td>
<td>2164 ± 433</td>
<td>4.1 ± 1.0</td>
</tr>
</tbody>
</table>

The very narrow range of registered ratios of $^{241}\text{Pu}/^{241}\text{Am}$ varies from 2.8 to 4.2, with an average of 3.7.

The date of the test is January 15, 1965, the theoretically calculated ratio of $^{241}\text{Pu}/^{241}\text{Am}$ is equal to 5.3 today with an average of 3.7 ± 1.0.

The purpose of the test - "Industrial nuclear explosions for peaceful purposes and the development of the technology of peaceful nuclear explosions." Again, it is logical to assume that a serial charge was used for the test. Estimated charge creation time is 1958-1959. i.e. charge lay in storage for about 7 years.

The "Combat Radioactive Substances" site.
The very narrow range of registered ratios of $^{241}\text{Pu}/^{241}\text{Am}$ varies from 0.6 to 1.5, with an average of 1.3. Table 3 presents the results of determining $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratio in soil samples taken at the "Combat Radioactive Substances" site.

Table 3. Results of determination of $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratios in soil samples taken on the site "Combat Radioactive Substances".

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{241}\text{Am}$, Bq/kg</th>
<th>$^{241}\text{Pu}$, Bq/kg</th>
<th>$^{241}\text{Pu}/^{241}\text{Am}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>БРВ-01</td>
<td>20 ± 1.6</td>
<td>29.8±6.0</td>
<td>1.5±0.4</td>
</tr>
<tr>
<td>БРВ-02</td>
<td>26 ± 1.8</td>
<td>45.2±9.0</td>
<td>1.7 ±0.4</td>
</tr>
<tr>
<td>БРВ-03</td>
<td>690 ± 57</td>
<td>448±90</td>
<td>0.6±0.2</td>
</tr>
</tbody>
</table>

Tests at the "CRS" site were conducted from 1953 to 1957, the range of theoretically calculated ratios of $^{241}\text{Pu}/^{241}\text{Am}$ today ranges from 2.4 to 4.7. However, in this case, it is difficult to theoretically calculate the $^{241}\text{Pu}/^{241}\text{Am}$ ratio. Contamination of this territory occurred due to sputtering of mixtures of combat radioactive substances made from spent reactor fuel after radiochemical separation of plutonium isotopes, therefore, containing a significant amount of $^{241}\text{Am}$ which accumulated in the reactor. This fact leads to a significant overestimation of the theoretically calculated range of relationships.
"Experimental field" and its zone of influence.

Directly on test sites in 90% of cases, the $^{241}\text{Pu}$ / $^{241}\text{Am}$ ratio lies in a rather narrow range from 1.3 to 3.9 with an average value of 2.4. Table 4 presents the results of determining $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratio in soil samples taken at the "Experimental field" site.

Table 4. Results of determination of $^{241}\text{Pu}$ and $^{241}\text{Am}$ and their ratios in soil samples taken on the site "Experimental field".

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{241}\text{Am}$, Bq/kg</th>
<th>$^{241}\text{Pu}$, Bq/kg</th>
<th>$^{241}\text{Pu}$/$^{241}\text{Am}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>ОП-01</td>
<td>480 ± 40</td>
<td>1680±330</td>
<td>3.5±1.0</td>
</tr>
<tr>
<td>ОП-07</td>
<td>124 ± 12</td>
<td>197±40</td>
<td>1.6±0.5</td>
</tr>
<tr>
<td>ОП-08</td>
<td>4730 ± 420</td>
<td>9120±1825</td>
<td>2.0±0.5</td>
</tr>
<tr>
<td>ОП-09</td>
<td>107 ± 8</td>
<td>542±110</td>
<td>5.0±1.5</td>
</tr>
<tr>
<td>ОП-10</td>
<td>44,4 ± 4,1</td>
<td>171±34</td>
<td>3.9±1.0</td>
</tr>
<tr>
<td>ОП-11</td>
<td>710 ± 140</td>
<td>920±184</td>
<td>1.3±0.3</td>
</tr>
<tr>
<td>ОП-12</td>
<td>61,1 ± 4,7</td>
<td>178±35</td>
<td>3.0±0.7</td>
</tr>
<tr>
<td>ОП-13</td>
<td>570 ± 40</td>
<td>1400±280</td>
<td>2.5±0.6</td>
</tr>
<tr>
<td>ОП-14</td>
<td>315 ± 28</td>
<td>617±123</td>
<td>2.0±0.5</td>
</tr>
</tbody>
</table>

Tests on the "Experimental Field" site were conducted from August 29, 1949 to January 24, 1962. Provided that the activity of $^{241}\text{Am}$ in the charge substance at the time of testing is minimal (for further calculations it is taken as zero), taking into account the law of radioactive decay, the range of theoretically calculated ratios of $^{241}\text{Pu}$ / $^{241}\text{Am}$ today varies from 2.4 to 4.7.

CONCLUSION

In most cases, at local sites, the $^{241}\text{Pu}$ / $^{241}\text{Am}$ ratio lies in a fairly narrow interval, which makes it possible to estimate the activity of $^{241}\text{Pu}$ from the results of $^{241}\text{Am}$ gamma-spectrometric analysis. The accuracy of estimating $^{241}\text{Pu}$ activity by this method will be about 50%, which in most cases is sufficient. However, this assessment method is not applicable to all STS test sites. Exceptions are the platforms "Combat Radioactive Substances" and "Degelen", because pollution on the "CRS" site has the character of local spots, with significantly different isotopic ratios, and on the "Degelen" site, the effect of redistribution along the stream channel is significantly manifested.

REFERENCES


FEATURES OF THE $^{90}$Sr DISTRIBUTION IN THE ABIOTIC COMPONENTS OF THE WATER ECOSYSTEMS OF THE LAKES OF CRIMEA

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ABSTRACT

A comparative study of the $^{90}$Sr distribution between the abiotic components of the lakes of Crimea with different levels of salinity from Yevpatoriyskaya (Lake Sasyk-Sivash, Lake Kyzyl-Yar) and Kerchenskaya (Lake Adzhigol, Lake Kuchuk-Adzhigol) groups of lakes was made. It is noted that the concentration of $^{90}$Sr in water and bottom sediments of Lake Adzhigol (150 ‰) is 3.2 and 7.4 times higher, respectively, than in Lake Kuchuk-Adzhigol (5 ‰). The concentration of $^{90}$Sr in water of Lake Sasyk-Sivash (280 ‰) is 20.8 times higher than in Lake Kyzyl-Yar (3.7 ‰), and in bottom sediments it is less than 7.5 times. It was determined that the level of $^{90}$Sr concentration in the ecosystems of the studied objects depends on the sources entry of this radionuclide, and the distribution of this radionuclide depends on the hydrochemical characteristics of the lakes, primarily on salinity. The increased salinity of the water environment in lakes reduces to 7.5 times the elimination of this radionuclide into bottom sediments.

KEYWORDS: $^{90}$Sr, salt lakes, water, bottom sediments, Crimea

1. INTRODUCTION

After the Chernobyl NPP accident in 1986, the maximum concentration of $^{90}$Sr entering the surface of Crimea and the Black Sea with atmospheric deposition was 98.1 Bq/m$^3$, and in 1987 the maximum concentration of this radionuclide that arrived by water was equal to 991.6 Bq/m$^3$ (Polikarpov et al., 2008). In the post-accident years, the radioecological situation in Crimea was determined by secondary radionuclide pollution, primarily $^{90}$Sr, with the runoff of water of the Dnieper river through the North-Crimean Canal (NCC), (Gulin et al., 2016). The first determinations of the content of the post-accident $^{90}$Sr in the water of the lakes of Crimea were performed in 2013 on the hypersaline lakes of the Perekopskaya group, and it was shown that the concentration of $^{90}$Sr from lake to lake significantly changed (Mirzoyeva et al., 2015). Presented in this paper, the results of the study are relevant, as they fill the gap in the study of the concentration and redistribution of $^{90}$Sr in aquatic ecosystems of salt lakes of Crimea. In 2018 for the lakes of the Kerch group Adzhigol and Kuchuk-Adzhigol, these radioecological studies were conducted for the first time in the history of the existence of these lakes.

The aim of this study was to identify peculiarities the current radioecological state of lakes with different salinity levels and differing from each other in hydrophysical properties.
2. MATERIAL AND METHOD

The work was carried out on the basis of sampling materials in expeditions of 2018. The studied objects of study are highlighted in squares in Figure 2.1.

![Fig. 2.1 Location of objects under study](image)

The determination of $^{90}$Sr in the objects under study was carried out by a radiochemical method. Beta - activity of $^{90}$Sr was measured by the Cherenkov radiation of its daughter radionuclide - $^{90}$Y on a low-background liquid scintillation analyzer «Quantulus-1220», the lower limit of the detected activity was 0.01-0.04 Bq∙kg$^{-1}$ sample, the relative error didn’t exceed 20% (Polikarpov et al., 2008; Mirzoyeva et al., 2015). 13 water samples and 60 sediment samples were collected and analyzed.

3. RESULTS AND DISCUSSION

According to previously obtained data (Mirzoyeva et al., 2018) and current results, the ranking of average concentrations of $^{90}$Sr in the water and bottom sediments of the studied lakes over the observation period 2016-2018 years is presented in fig. 3.1(a) and fig. 3.1(b), respectively.

![Fig. 3.1 Distribution of $^{90}$Sr concentration in water (a) and in bottom sediments (b) of the studied lakes](image)

It is shown (Fig. 3.1) that one of the highest concentrations of $^{90}$Sr is observed in the water of the lakes Sasyk-Sivash and Adzhigol (26.2 Bq/m$^3$ and 107.3 Bq/m$^3$), and the lowest in the water of the lakes Kyzyl-Yar and Kuchuk-Adzhigol (10.1 Bq/m$^3$ and 33.1 Bq/m$^3$). The concentration of $^{90}$Sr in the water of the Black sea by 2018 is equal 10.2 Bq/m$^3$. From fig.3.1...
(b) it is clear that one of the highest concentrations of $^{90}$Sr in bottom sediments are data obtained in lakes Adzhigol and Kyzyl-Yar (18.9 Bq/kg and 3.1 Bq/kg), while the lowest - in lakes Kuchuk-Adzhigol and Sasyk-Sivash (2.6 Bq/kg and 6.6 Bq/kg). It was noted that the concentration of $^{90}$Sr in water and bottom sediments of Lake Adzhigol (150 ‰) was 3.2 and 7.4 times higher, respectively, compared to that of Lake Kuchuk-Adzhigol (5 ‰), Kerchenskaya group of lakes. The concentration of $^{90}$Sr in water of Lake Sasyk-Sivash (280 ‰) was 20.8 times higher and bottom sediments was 7.5 times less, compared to that of Lake Kyzyl-Yar (3.7 ‰), Yevpatoriyskaya group of lakes. So, there is a direct correlation of the concentration of $^{90}$Sr from the salinity in water of all four of the studied objects (Fig. 3.2 (a, b)).

![Fig. 3.2 $^{90}$Sr in water (a, b) and in bottom sediments (c, d) of the studied lakes](image)

Figure 3.2 shows the inverse dependence of the concentration of $^{90}$Sr on growth of the salinity in the water of lakes, while for the lakes of the Kerchenskaya group this trend wasn't confirmed which may be due to the different rates of sedimentation in these lakes.

From the obtained results it can be seen that the sources of income $^{90}$Sr for the lake Kuchuk-Adzhigol, in the absence of communication with the NCC, were atmospheric depositions (Fig. 3.3).

![Fig. 3.3 Restored concentration $^{90}$Sr in water of studied objects on 1986](image)

For the lakes Sasyk-Sivash and Adzhigol, the largest contribution was made by the waterway, the secondary route of entry is probably the Black sea, due to the difference in salinity and in hydrophysical properties of objects of investigation. Until 2014, the Kyzyl-Yar Lake was directly connected with the NCC, which contributed to the chronic secondary flow of the post-accident $^{90}$Sr into the ecosystem of this water body, however, the biogeochemical
features of this water body have a significant impact on the processes of migration and accumulation of the radionuclide.

4. CONCLUSION

For the first time radioecological studies were performed on the content and distribution of $^{90}\text{Sr}$ in the abiotic components of two lakes of Crimea of the Kerchenskaya group Adzhigol and Kuchuk-Adzgigol. In lakes Sasyk-Sivash, Adzhigol and Kyzyl-Yar, Kuchuk-Adzhigol, both the highest and lowest concentrations of $^{90}\text{Sr}$ in water and bottom sediments are observed, which makes these studies relevant and allow for a comparative analysis of lakes similar in their physicochemical characteristics.

The sources of $^{90}\text{Sr}$ entry into the lakes of the Kerchenskaya and Yevpatoriyanskaya groups were determined: only from atmospheric precipitation in 1986 after the Chernobyl NPP accident (Kuchuk-Adzhigol lake), predominance of secondary inflow from the Black Sea (Sasyk-Sivash and Adzhigol lakes), inflow with the Dnieper river waters through the NCC (Kyzyl-Yar lake).

A direct dependence of the concentration of dissolved $^{90}\text{Sr}$ on the salinity level of the investigated lakes was revealed.

ACKNOWLEDGEMENTS

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5. REFERENCES


INFLUENCE OF CONTAMINATED SEDIMENTS IN SCHENKINO RESERVOIR ON $^{137}$Cs CONCENTRATION IN FISH

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ABSTRACT

The radionuclides content in the Shchekino reservoir (Tula region) fish was investigated. The results showed that the level of specific activity in the muscles of fish is 1-2 Bq/kg, which is much lower than the existing limits values (130 Bq/kg). The water physicochemical properties and specific activity of $^{137}$Cs of the reservoir were determined. Reservoirs conditions are comfortable for fish living and productivity. The reason for the content decrease in water and fish living in the Shchekino reservoir, in addition to the $^{137}$Cs half-life over the past 30 years after the accident, was the relatively clean river sediments accumulation at the bottom of the reservoir. This led to the most contaminated sediment layer burial after the accident time.

KEYWORDS: $^{137}$Cs, freshwater reservoirs, Chernobyl pollution, radionuclides in fish, radiation safety standards.

1. INTRODUCTION

The Chernobyl accident radionuclide fallout affected huge areas in Europe. Modern safety nuclear power plants system is well developed, however, as the accident at Fukushima-1 NPP on March 11, 2011 showed, it is very difficult to foresee all emergency situations possible causes and there is always a risk of their occurrence. Hydrobionts are a food chain component and are actively involved in the radionuclides migration in reservoirs and streams. Radioactive cesium is a nuclear reactions fission product, which has been included in the all living organisms natural habitat. Changes in the cesium concentrations in reservoirs are largely determined by adsorption and chemical interactions exchange with the surface of suspensions and bottom sediments. River suspended sediments can be source of secondary water radioactive contamination as a result of desorption processes caused by external environmental conditions changes. However, biota can have a significant impact on the redistribution of radionuclides in the environment. Biotic interactions can, for example, by changing alkaline-acid conditions, help to obtain soluble ligands or new biomineral phases (Newsome, 2014).

The Shchekino reservoir is located in Tula region within the “Plavskiy radioactive spot” impact zone, which is characterized the highest initial contamination level on the long distance from Chernobyl NPP. The level of contamination of soil exceed 185 kBq/m$^2$ after initial fallout in 1986 in the reservoir catchment. Maximum specific activity of Cs-137 in bottom sediments was about 4500 Bq/kg in 1986.

The goal of our study is to evaluate the $^{137}$Cs transfer from catchment area for the 32 years after accident and to determine the influence of $^{137}$Cs sedimentation in reservoir on the fish.
2. MATERIAL AND METHOD

Field investigations were undertaken in low water periods (January and July 2018). In February 2018 four bottom sediment cores were taken in the typical locations of the upper half of reservoir, which were selected using results of georadar survey of reservoir bottom along the few cross section profiles. 17 samples of fish were collected (1 carp (Hypophthalmichthys), 12 crucian carp (Carassius), 4 roach (Rutilus rutilus) from reservoir in July 2018. When fishing was taken, the catch season, fish age, length, weight, gender, exact fish name and sampling coordinates were entertained. After catching, the fish were washed to reduce the likelihood of sediment particles containing cesium, then crushed, packed in tight plastic containers and weighed. In order to identify $^{137}$Cs correctly, fish was gutted and only parts which could be eaten was selected for analysis. In some cases, the, fish gutted, the internal parts and muscles were analyzed separately. Caviar was separated from several fish, the carp was divided into organs (gills, head, liver, tail part). $^{137}$Cs activity in samples of sediment and fish was measured using a semiconductor gamma spectrometer with a HPGe detector. In addition, hydrological and hydrochemical investigations were carried out in low-flow periods. The electrical conductivity and water pH, the concentration of major ions, biogenic elements, as well as indirect indicators of the organic substances content (chromaticity, permanganate oxidation (PO), chemical oxygen consumption (COD) were determined in reservoir water samples.

3. RESULTS AND DISCUSSION

The reservoir water is characterized by the moderate mineralization belong to the hydrocarbonate class, the group of calcium and magnesium and it is slightly alkaline (pH - 7.6 -8.5). Based on the interpretation of $^{137}$Cs vertical distribution in the bottom sediments, the $^{137}$Cs concentration dynamics changes in sediment yield of the Upa River over 1986-2018 were restored. During the post-Chernobyl period, radionuclides concentration in sediments reduced considerably. Therefore, intensive siltation resulted into burial of sediment layers with maximum concentrations of $^{137}$Cs. The total value of COD in summer varied in the range of 8-25 mgO/l, which is lower than the established value of limits (30 mgO/l). More than 50% was an organic matter suspended form. The potassium content is low (0.5-1.2 mg/l). All of the hydrochemical parameters in general are comfortable for fish living and reproductive ability. The $^{137}$Cs specific activity of some fish samples presents in Table 1.

Table 1. Characteristics of studies fish and $^{137}$Cs specific activity in some Shchekino reservoir samples.

<table>
<thead>
<tr>
<th>Fish type</th>
<th>Gender</th>
<th>Length, см</th>
<th>Weight, g</th>
<th>A, Bq/kg</th>
<th>Uncertainties, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crucian carp</td>
<td>F</td>
<td>23,5</td>
<td>226,7</td>
<td>0,45</td>
<td>1,0</td>
</tr>
<tr>
<td>Crucian carp caviar</td>
<td>F</td>
<td>-</td>
<td>15,4</td>
<td>1,91</td>
<td>5,0</td>
</tr>
<tr>
<td>Roach</td>
<td>M</td>
<td>18,5</td>
<td>68,2</td>
<td>1,61</td>
<td>12</td>
</tr>
<tr>
<td>Silver carp (muscles from the tail part)</td>
<td>M</td>
<td>59,0</td>
<td>56,1</td>
<td>1,6</td>
<td>17</td>
</tr>
</tbody>
</table>
Exploration of fish showed that the level of $^{137}$Cs activity in the muscles is much lower than the existing maximum permissible level – 130 Bq/kg. The obtained data allow us to conclude that the current state of the Shchekino reservoir fish is satisfactory assessed.

After the Chernobyl and Fukushima nuclear power plants accidents the correlations between $^{137}$Cs concentrations in lake sediments and fish has been established in a number of studies. A high concentration of $^{137}$Cs in porous (light) sediments is associated with higher concentrations of $^{137}$Cs in zooplankton and aquatic insects that fish feeds, which leads to a higher concentration of $^{137}$Cs in fish itself (Håkanson, 1989; Jagne, 1998; Fukushima, 2014). A high sedimentation rate can lead to more rapid $^{137}$Cs accumulation in sediments, which will affect the radionuclides accumulation by fish. This situation described by the example of the typhoon effects (Nagao, 2013). Also, it was determined a longer biological half-life of $^{137}$Cs from fish in lakes in the Fukushima area with a slow water exchange, compared with lakes with fast water exchange. For Shchekino reservoir, a sufficiently fast water exchange was established, which positively affects the self-cleaning of the object from $^{137}$Cs. Table 2 presents modern specific activities in bottom sediments and fish muscles from lakes around Fukushima-1.

<table>
<thead>
<tr>
<th>Species</th>
<th>Lake</th>
<th>$^{137}$Cs activity in sediments</th>
<th>A, Bq/kg (fish muscles)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crucian carp</td>
<td>Inavashiro</td>
<td>$1.2 \times 10^{2}$</td>
<td>88</td>
</tr>
<tr>
<td>Crucian carp</td>
<td>Akimoto</td>
<td>$3.2 \times 10^{2}$</td>
<td>61</td>
</tr>
<tr>
<td>Crucian carp</td>
<td>Numazava</td>
<td>$2.9 \times 10^{2}$</td>
<td>150</td>
</tr>
<tr>
<td>Silver carp</td>
<td>Tagokura</td>
<td>$1.2 \times 10^{2}$</td>
<td>&lt;MDA</td>
</tr>
</tbody>
</table>

If we compare this type of objects in the territories affected by the Chernobyl impact and the Fukushima-1 nuclear power plant accident it is clear that redeposition and burial of bottom sediments contaminated layers in Japan reservoirs accelerate through typhoon season. In addition, the large areas around the Fukushima-1 were decontaminated recently, which also reduces $^{137}$Cs concentration in sediment entering the reservoirs.

4. CONCLUSION

According to the Russian standard (SanPin 2.6.1.2523-09 “Radiation Safety Standards (NRB-99/2009)”, the maximum allowable specific $^{137}$Cs activities for fish are 130 Bq/kg. The results of determination of $^{137}$Cs in fish of the Shchekino reservoir are significantly below the limits. Investigated fish is safe for food. This is due to the fact that despite the location of the reservoir in the initially highly contaminated Chernobyl fallout zone, the time elapsed since the accident (32 years) and suspended sediment promoted to burial of the most contaminated bottom sediment horizon, which may be the source of radioactive substances entering the fish. In the territories affected by Fukushima-1 nuclear power plant radionuclide fallout burial processes of contaminated horizons of bottom sediments are going faster because of the regular high water floods associated with typhoons.
ACKNOWLEDGEMENTS

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5. REFERENCES


NATURE OF RADIOACTIVE CONTAMINATION IN SOILS OF THE PINE FOREST IN THE TERRITORY ADJACENT TO SEMIPALATINSK TEST SITE

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ABSTRACT
The paper provides specific activity values for $^{137}$Cs, $^{90}$Sr, $^{241}$Am, $^{239+240}$Pu artificial radionuclides in pine forest at the territory, adjacent to Semipalatinsk Test Site. Pattern of these radionuclides distribution along the soil profile was considered. The maximum concentration of $^{241}$Am, $^{137}$Cs and $^{239+240}$Pu was registered in the top 5-cm thick layer, and $^{90}$Sr is uniformly distributed up to the depth of 20 cm. In general, researched artificial radionuclides by the level of their activity can be represented by the row as follows: $^{90}$Sr $>$ $^{239+240}$Pu $>$ $^{241}$Am $>$ $^{137}$Cs.

KEYWORDS: Semipalatinsk Test Site (STS), artificial radionuclides, cesium ($^{137}$Cs), strontium ($^{90}$Sr), americium ($^{241}$Am), plutonium ($^{239+240}$Pu), soil.

1. INTRODUCTION
At the present time radioactively contaminated territories near Itysh river are still of interest. This contamination was formed 60 years ago after radioactive fallout plume have passed this area as the result of a nuclear test conducted on the 29th of August, 1949 at Semipalatinsk Test Site (STS). Parameters of radionuclides redistribution among the individual components of forest ecosystems at this territory can be used to predict radioecological situation development. Also, obtained data can be used as input parameters for the models, used to describe radioactive substances behavior and to assess the risk of contamination for forest ecosystem components long after radioactive contamination. Scientific novelty is also explained by transuranium radionuclides ($^{241}$Am, $^{239+240}$Pu) at the territory researched, because the basic data available globally were mainly obtained for $^{137}$Cs and $^{90}$Sr.

This research was aimed at determining levels and character of soil contamination with artificial radionuclides ($^{241}$Am, $^{137}$Cs, $^{90}$Sr, $^{239+240}$Pu) in the forest ecosystem at the territory adjacent to the STS.

2. MATERIAL AND METHOD
For the purpose of research, the areas with presumably elevated level of radioactive contamination were chosen. For this purpose, $\beta$-particles flux density and equivalent dose rate (EDR) were measured. In total, 6 research areas were equipped. At each of research areas a soil profile was laid out. To assess the character of radionuclides migration along the soil profile soil samples were collected in 5 cm intervals to the depth of 25 cm.
Before the radionuclide analysis, soil samples were dried in drying cabinets at the temperature of 50-60°C to air-dry state. After removal of coarse rocks and inclusions (plant residues) dried samples were weighted using technical scales. Next, the whole sample mass was carefully mixed, with gradual mashing in porcelain mortar using a beater and sieved through 1 mm mesh. To check the completeness of sieving, each mesh was shaken over a sheet of paper.

Specific activity of $^{137}$Cs and $^{241}$Am was determined using Canberra GX-2020 γ-spectrometer, while $^{90}$Sr and $^{239+240}$Pu were determined by means of radiochemical extraction with further measurement with TRI-CARB 2900 TR β-spectrometer and Canberra α-spectrometer (mod. 7401) respectively. Detection limit was 0.1 Bq/kg. Measurement error for $^{137}$Cs and $^{241}$Am did not exceed 10-20%, while for $^{90}$Sr and $^{239+240}$Pu - 15-25% and 30% respectively.

3. RESULTS AND DISCUSSION

Upon the results of analyses, the highest specific activity values at the researched territory are typical for $^{241}$Am and $^{239+240}$Pu transuranium radionuclides (up to 64 Bq/kg and 75 Bq/kg respectively). Specific activity of $^{137}$Cs in most of the cases is below the detection limit of equipment and methodology used, the maximum value was registered at the site 6 (75 Bq/kg). Quantitative values of $^{90}$Sr specific activity was found in all the collected samples, however the maximum values do not exceed 8,3 Bq/kg.

The average percentage of $^{137}$Cs, $^{241}$Am and $^{239+240}$Pu in 0-5 cm layer is 81,3 %, 50,2% and 60,7 % of its total specific activity in soil vertical profile respectively. At the depth of up to 10 cm 98,9 % of $^{137}$Cs specific activity, 69,3% and 75,2 % of $^{241}$Am and $^{239+240}$Pu specific activity respectively was registered. The percentage of $^{90}$Sr in 0-5 cm layer is 25,9 %, and further, up to 20 cm deep the radionuclide is distributed virtually uniformly. At the depth of up to 10 cm, only 42,5% of the total specific activity found in the researched vertical soil profile was registered. High mobility of $^{90}$Sr in this case can be caused by its higher migration ability due to good solubility.

4. CONCLUSION

As the result of researches, conducted in pine forest at the territory, adjacent to the STS, increased concentrations of all the radionuclides considered ($^{241}$Am, $^{90}$Sr, $^{137}$Cs, $^{239+240}$Pu) were registered. The highest specific activity values were registered for $^{241}$Am (up to 64 Bq/kg) and $^{239+240}$Pu (up to 75 Bq/kg) transuranium radionuclides. Maximum concentrations of $^{241}$Am, $^{137}$Cs and $^{239+240}$Pu were registered in the top 5-cm layer, while $^{90}$Sr is uniformly distributed up to the depth of 20 cm. In general, researched artificial radionuclides by their level of mobility in vertical soil profile can be arranged in sequence as follows: $^{90}$Sr > $^{239+240}$Pu > $^{241}$Am > $^{137}$Cs.

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Yoschenko V. et al. Radiocesium distribution and fluxes in the typical Cryptomeria japonica forest at the late stage after the accident at Fukushima Dai-Ichi Nuclear Power Plant /

The concentration of the anthropogenic radionuclide $^{137}\text{Cs}$ was studied in the abiotic components of the ecosystems of the salt lakes of Crimean region: lake Sasyk-Sivash and lake Kyzyl-Yar in 2016-2018. It was determined that the salinity of water is the main geochemical factor affecting on the distribution of $^{137}\text{Cs}$ between the abiotic components of the studied lakes.

**KEYWORDS:** Crimea, salt lakes, $^{137}\text{Cs}$, water, bottom sediments.

**1. INTRODUCTION**

Crimean salt lakes are of balneological, industrial and recreational importance. They are divided into the following major groups: Perekopskaya, Kerchenskaya, Tarkhankutskaya and Evpatoriiskaya (Aizenberg and Kaganer, 1966). The main source of technogenic radionuclides in salt lakes was atmospheric fallout after the accident at the Chernobyl nuclear power plant in 1986. According to researchers, about 4 PBq of $^{137}\text{Cs}$ fall out on the Black Sea, including surface water bodies of the Crimean Peninsula, after Chernobyl accident (Polikarpov et al., 2008). An additional source of radioactive pollution of Crimean salt lakes has been shown to be the input through the North Crimean Canal of Dnieper water, carrying high concentrations of Chernobyl radionuclides. About 674 GBq of $^{137}\text{Cs}$ flowed to the North Crimean canal from the Kakhov reservoir for the period 1986-1995 (Gulin et al., 2016). The salt Lake Sasyk-Sivash and Lake Kyzyl-Yar, relating to the group of Yevpatoria of the Crimean peninsula, was investigated for the content of the anthropogenic radionuclide $^{137}\text{Cs}$ as an important factor of water pollution. The choice of objects of research was related to the opposite mechanisms of $^{137}\text{Cs}$ distribution in the abiotic components of these lakes despite the close geographical location of these reservoirs, but different sources of anthropogenic radionuclides. The purpose of work was determination of $^{137}\text{Cs}$ distribution regularities between water and bottom sediments of lakes Sasyk-Sivash and Kyzyl-Yar during 2016-2018.

**2. MATERIAL AND METHOD**

Samples of water and bottom sediments of studied lakes were collected at the period 2016-2018. The scheme of location of the lakes is given in Fig. 1. To determine the content of $^{137}\text{Cs}$ in water samples (50-100 l) a method based on $^{137}\text{Cs}$ concentration by flow sorption in two series-connected adsorbers was used, followed by the measurement of the content of $^{137}\text{Cs}$.
$^{137}$Cs by gamma radiation of the daughter radionuclide $^{137m}$Ba (Gulin et al., 2015). The radiometric measurements were carried out using NaI(Tl) well-type gamma spectrometer COMPUGAMMA (LKB Wallac, Finland) subject to radioactive background and efficiency of $^{137}$Cs detection. The efficiency of sorption of the dissolved $^{137}$Cs was calculated by comparison of activities on the first and second serial adsorbers. After that, the concentration of $^{137}$Cs in the water sample was calculated (Gulin et al., 2015).

Bottom sediments of salt lakes were taken using an acrylic tube with an internal diameter of 58 mm. The columns were cut into layers 1 and 5 cm thick using a piston extruder (Gulin and Gulina, 2009). Then the precipitates were dried at a temperature of 100 °C, ground in a porcelain mortar and weighed. The ground samples analyzed for semiconductor gamma detector based on a crystal of ultrapure germanium (Canberra, USA). The $^{137}$Cs content in the sample was calculated on the dry weight of the sediments after correction with the weight of salts dissolved in pore water and taking into account the radioactive background (Stokozov and Gulin, 2008).

![Fig. 1. Map-scheme of sampling stations](image)

3. RESULTS AND DISCUSSION

It was determined that the primary source of $^{137}$Cs in the lake Kyzyl-Yar and lake Sasyk-Sivash were atmospheric deposition after the Chernobyl accident in 1986. The maximum $^{137}$Cs concentration in Black Sea water after accident was 815 Bq/m$^3$ (Polikarpov et al., 1992), which exceeded the levels of $^{137}$Cs in the lakes according to radioactive decay.

Three-year dynamics of $^{137}$Cs content in water and bottom sediments of the studied lakes was obtained for the first time (table 1). The level of $^{137}$Cs in the water of Lake Sasyk-Sivash for the period 2016-2018 changed from 83 to 142 Bq/m$^3$, but in the bottom sediments $^{137}$Cs was not found. The opposite trend was observed in Lake Kyzyl-Yar. The level of $^{137}$Cs in water changed from 0.75 to 2.90 Bq/m$^3$, and in bottom sediments changed from 15 Bq/kg.
to low detection limit. Mechanism of distribution of $^{137}$Cs between components the components of lakes depends on the salinity of reservoirs. Thus, in Lake Sasyk-Sivash, which has a very high salinity (average 290‰) the concentration of $^{137}$Cs in water was maximum. And in the Lake Kyzyl-Yar during the study period, salinity increased from 3.5 to 7.0‰, while the content of $^{137}$Cs in water increased almost three times, and in bottom sediments decreased to minimum value. Low salinity of lake Kyzyl-Yar was caused by discharge excess fresh Dnieper water from the Meigornoe reservoir flowing through the North Crimean Canal (Shadrin et al., 2018) before 2014. Therefore, the $^{137}$Cs elimination from the aquatic environment occurred primarily due to biogeochemical processes. In this case, the decrease of $^{137}$Cs concentration was mainly due to radioactive decay, and high salinity keeps $^{137}$Cs in a dissolved form. Therefore, the salinity of the lakes directly affects the accumulation of $^{137}$Cs of aquatic environment.

According to the results (table 1) it is shown that the concentration of $^{137}$Cs in bottom sediments of studied lakes is significantly lower than in the Black Sea (Fig.5). For example, the average concentration of $^{137}$Cs in the surface layer of bottom sediments of Sevastopol Bay (The Black Sea) is 95.5 Bq/kg (Egorov et al., 2018).

Table 1. The concentration of $^{137}$Cs in the water and bottom sediments of salt lakes.

<table>
<thead>
<tr>
<th>Lake</th>
<th>Concentration in the water, Bq/m$^3$</th>
<th>Concentration in the bottom sediments, Bq/kg (dry weight)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kyzyl-Yar</td>
<td>0.8±0.04</td>
<td>1.7±0.1</td>
</tr>
<tr>
<td>Sasyk-Sivash</td>
<td>95.9±8.2</td>
<td>95.9±8.2</td>
</tr>
</tbody>
</table>

In 2016-2018 concentration of $^{137}$Cs in water of all investigated salt lakes did not exceed levels the maximum permissible concentration for $^{137}$Cs in drinking water according to radiation safety standards of the Russian Federation (RSS–99/2009).  

4. CONCLUSION

A comparative assessment of the $^{137}$Cs content in the salt lakes of Crimea region was obtained. The main sources of technogenic radionuclides in the lakes was atmospheric fallout after Chernobyl accident. The dynamics of $^{137}$Cs content in the Evpatoriiskaya group of salt lakes was obtained. The factors of $^{137}$Cs elimination from the aquatic environment of lakes of Evpatoriiskaya group is biogeochemical processes and radioactive decay. It was determined, that salinity directly affects the accumulation of $^{137}$Cs of lake water and it is an important biogeochemical factor of migration capacity of $^{137}$Cs between the components of ecosystems of saline lakes of Crimea region. And the proof of that is the opposite mechanisms of distribution of man-made radionuclide $^{137}$Cs between the abiotic components of the almost
freshwater Lake Kyzyl-Yar and hypersaline Lake Sasyk-Sivash related to Evpatoriiskaya group of Crimean salt lakes.

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REFERENCES


NATURAL RADIONUCLIDE $^{210}$Po IN AQUATIC ECOSYSTEMS OF SALT LAKES OF CRIMEA

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ABSTRACT

A monitoring radioecological study regards the content of natural radionuclide $^{210}$Po in aquatic ecosystems of salt lakes in Crimea was carried out for the first time in the history of the existence of these lakes. Additional, besides of atmospheric, sources of $^{210}$Po in the water of Tobechikskoe and Chokrakskoe lakes (Kerchenskaya group of salt lakes of Crimea) were identified. The average concentrations of dissolved $^{210}$Po in the water of salt lakes were 0.9–327.1 times higher than its content in the Black Sea (0.7 Bq·m$^{-3}$). A significant trend in a decrease of Accumulation Coefficient (AC) of $^{210}$Po by suspended matter with an increase in the salinity of the aquatic environment of the saline lakes of Crimea was revealed. The $^{210}$Po concentrations in the bottom sediments of salt lakes of Crimea were in the range of values observed in the bottom sediments of the Black Sea. Artemia spp. from the saline lakes of the Crimea accumulated significant amounts of $^{210}$Po (up to 200 Bq·kg$^{-1}$ W.W.), while the concentrations of this radionuclide in its cysts were an order of magnitude lower. The absorbed dose rate of $^{210}$Po $\alpha$-radiation received by Artemia spp. (adult animals), is more than 60 times lower than the dose rate recommended by the IAEA as acceptable for biota (IAEA, 1992).

KEYWORDS: $^{210}$Po, salt lakes of Crimea, water, bottom sediments, hydrobionts

1. INTRODUCTION

By now salt lakes remain the least studied in relation to the behavior of $^{210}$Po (Yadav and Sarin, 2009). The actuality of this radioecological study concerning the content of $^{210}$Po in the ecosystems of saline lakes of Crimea is that in the history of all saline lakes of Crimea and for the selected objects such studies were carried out for the first time.

The purpose of this work is to determine the biogeochemical features of the formation of the modern radioecological state of the Crimean salt lakes with respect to the $^{210}$Po accumulated by different components of their aquatic ecosystems.

In accordance with the assigned goal, the following tasks were solved: to determine of $^{210}$Po concentrations in components of the ecosystems of the salt lakes of the Crimea; to carry out a comparative analysis of the $^{210}$Po content in water, aquatic organisms, bottom sediments of salt lakes of the Crimea and the Black Sea ecosystems close to their location; to assess of the biogeochemical characteristics of the aquatic ecosystems in the transport and removal of $^{210}$Po from the water column of the salt lakes of the Crimea; calculate the radiation dose
received by different hydrobionts of the salt lakes of the Crimea from ionizing radiation of $^{210}$Po.

2. MATERIAL AND METHOD

The materials for this study were sampled in several salt lakes of Crimea during 2016-18 (Fig. 2.1).

![Location of the investigated salt lakes in the Crimea](image)

**Fig. 2.1.** Location of the investigated salt lakes in the Crimea (1-6 - control sampling points in the Black Sea).

It was sampled and analyzed 19 samples of water, 10 samples of suspended matter, 29 samples of bottom sediments, 6 samples of hydrobionts (Crustacea (*Artemia* spp. (adults and cysts)).

$^{210}$Po activity concentration in the samples was calculated according to the methods described in (EPA, 1984). The permissible error of $^{210}$Po determination (Lazorenko, Polikarpov, 2008) did not exceed 20% for water, and 10% for bottom sediments and hydrobionts.

To calculate the absorbed dose rates formed by the radiation of $^{210}$Po alpha particles in lives organisms, the approaches and criteria described in (Lazorenko, Polikarpov, 2008).

3. RESULTS AND DISCUSSION

It was obtained (Fig. 3.1) that the average concentrations of $^{210}$Po in the filtered water samples of salt lakes as a whole were 0.9-327.1 times higher than its content in the Black Sea. The increased content of $^{210}$Po in lakes of the Kerchenskaya group (Fig. 3.1(a)) is possibly due to the presence of oil fields on the Kerch Peninsula. It is known, that elevated concentrations of natural radionuclides of a number of uranium and thorium are observed in the ground and surface waters of such regions, including polonium, which is formed during the decomposition of gaseous $^{222}$Rn, penetrating into the surface water from the oil-bearing horizons.
Fig. 3.1. $^{210}$Po in dissolved water (a) and bottom sediments (b) of the salt lakes of Crimea, where lakes: a) 1 – Moynakskoe, 2 – Kyzyl-Yar, 3 – Bakalskoe, 4 – Dzharylgach, 5 – Krasnoe, 6 – Kirleutskoe, 7 – Aktashskoe, 8 – Tobechikskoe, 9 – Chokrakskoe; b) 1 – Krasnoe, 2 – Kiyatskoe, 3 – Kirleutskoe, 4 – Bakalskoe, 5 – Dzharylgach, 6 – Sasyk-Sivash, 7 – Kyzyl-Yar, 8 – Moynakskoe, 9 – Aktashskoe, 10 – Chokrakskoe, 11 – Tobechikskoe.

Concentrations of $^{210}$Po in the bottom sediments of salt lakes of Crimea (Fig. 3.1(b)) were in the range of values noted in the bottom sediments of the Black Sea (from 14 Bq·kg$^{-1}$ D.W. to 242 Bq·kg$^{-1}$ D.W. (own data), on average - 140 Bq·kg$^{-1}$ D.W.). The Accumulation Coefficients (AC) of $^{210}$Po of the bottom sediments of salt lakes of Crimea varied in the range from n·10$^2$ to n·10$^4$. Lowest coefficients were determined for the Kerchenskaya group (eastern part of Crimea), and the largest - for the lakes of the Tarkhankutskaya and Yevpatoriyskaya group of lakes (northern-western part and western part of Crimea).

It was noted that Artemia (adults animals) from the Lake Dzharylgach accumulated significant amounts of $^{210}$Po (up to 200 Bq·kg$^{-1}$ W.W.) while the concentrations of this radionuclide in its cysts were an order of magnitude lower (Fig. 3.2(a)).

Fig. 3.2. $^{210}$Po in hydrobionts (a) and its AC of suspended matter depending on the salinity of water (b)

This is explained by the fact that the intake of polonium into animal organisms occurs only by food (Heyraud, Cherry, 1979), therefore, the concentrations of $^{210}$Po in cysts were insignificant.
A tendency towards a decrease of Accumulation coefficients (AC) of $^{210}$Po by the plankton of salt lakes (as well as for suspended matter) with increasing salinity of water was observed (Fig. 3.2(b)).

It was found that the absorbed dose rate from $^{210}$Po α-radiation obtained by Artemia (adult animals) is almost 2 times the maximum value determined for the Black Sea mussel *Mytilus galloprovincialis* (Lazorenko, Polikarpov, 2008). At the same time, the dose commitment on Artemia (adults animals) was more than 60 times lower than the dose rate recommended by the IAEA as permissible for biota (IAEA, 1992).

4. CONCLUSION

The dependence of the concentration of dissolved $^{210}$Po on the level of salinity in the studied lakes was not observed. The reliable trend of decrease of Accumulation Coefficients of $^{210}$Po by the suspended matter with an increase in the salinity of the water of the salt lakes of Crimea was determined. The dose commitment on Artemia (adult animals) was more than 60 times lower than the dose rate recommended by the IAEA as permissible for biota. The obtained results of the study allow to evaluate the previously unexplored effect of the radiation of alpha-particles of $^{210}$Po on the quality of the aquatic environment, hydrobionts, and to determine also the role of living and abiotic components in the transport, migration and elimination of this radionuclide in the ecosystems salt lakes of Crimea.

ACKNOWLEDGEMENTS

The results and conclusions in this article are based on the new data, which were obtained within the framework and at support by the Russian Scientific Foundation, grant № 18-16-00001.

5. REFERENCES


PREPARATION OF CHITOSAN-ALGINATE COMPOSITE HYDROGEL (CACH) FOR Sr (II) REMOVAL FROM AQUEOUS SOLUTION AND EVALUATION OF ADSORPTION PERFORMANCE

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ABSTRACT

This work reports the adsorption of Strontium (Sr (II)) ions from aqueous solutions onto Chitosan-Alginate Composite Hydrogel (CACH) was synthesized by dropwise method. The structural properties of CACH composites were investigated by scanning electron microscopy (SEM) and Fourier transformation infrared spectroscopy (FT-IR). Adsorption of Sr (II) ions onto CACH from aqueous solution was investigated in the batch experiment as a function of Sr (II) ion concentration, pH of the solution, contact time and temperature. In order to determine the adsorption characteristics, Langmuir, Freundlich, and Dubinin–Radushkevich adsorption isotherms were applied to the adsorption data. The thermodynamic parameters enthalpy, entropy and variation of Gibbs free energy were calculated from the slope and intercept of ln Kd vs. 1/T plots.

KEYWORDS: Sr (II), adsorption, adsorbent, chitosan, alginate, hydrogel.

1. INTRODUCTION

In the past four decades, the health problem because of the radioactive pollution have grown considerably. Radioactive contamination of foodstuff and water sources results is accumulation of elements such as Sr-90 and Cs-137 in nature, which are not excrete easily in the body. That is why; the study of the purification of water from these radioisotopes is very important and work is still going on. Some of these works for example; A facile synthesis of hydroxyapatite for effective removal Sr (II) ion (Xia et al., 2019); Removal of Sr (II) from aqueous solutions using scallop shell powder (Mihara et al., 2019); Removal of Sr (II) from aqueous solutions by acrylamide modified attapulgite (Zuo et al., 2019); Preparation and characterization of magnetic graphene oxide nanocomposite (GO-Fe3O4) for removal of Sr (II) and cesium from aqueous solutions (Yusan et al., 2018); Adsorptive removal of Sr (II) using macroporous poly (AGE-co-EGDMA) beads modified with resorcin[4]arene (Mulani et al., 2019); Surface functionalized diatomaceous earth for effective adsorption of Sr (II) from aqueous solution (Dhanapal, Ravindran and Selvakumar, 2019); Cobalt and Sr (II) sorption by moss biosorbent : Modeling of single and binary metal systems (Mare et al., 2011); Applied Surface Science Highly efficient capturing and adsorption of cesium and Sr (II) ions from aqueous solution by porous organic cage : A combined experimental and theoretical study (Ghalami, Ghoulipour and Khanchi, 2019).

Chitosan has a good hydro stability and good adsorbent for aqueous ions thanks to high surface area. Today chitosan and calcium alginate composite is already synthesized and properties investigated by Meng He (He et al., 2017). The alginic acid and alginates are thus likely to be potentially useful adsorbents for the removal of radionuclides from waste
solutions. However, a limited amount of information is available concerning the uptake behavior of radionuclides on these polymer gels (Gok and Aytas, 2009).

Because of the CACH exhibited good architectural stability. Sr (II) ions could adsorbed and stuck on the layer surface of CACH. This study aims the feasibility of using chitosan-alginate composite beads for the removal of strontium ions from aqueous solutions.

2. MATERIAL AND METHOD

The reagents used in the experiment were of analytical reagent (AR) grade. A solution of 1000 mg/L of Sr (II) was prepared from SrCl$_2$.6H$_2$O (Merck) by dissolving the salt in deionized distilled water. Sr (II) concentrations were determined by inductively coupled plasma optical emission spectrometry (Perkin-Elmer Optima 2000 DV ICP–OES). The adsorption experiments studied by batch technique.

Calcium alginate beads prepared by dropwise addition of 2% of viscous sodium alginate solution (w/v) to 0.5 mol/L CaCl$_2$ solution. Then 0.03 g chitosan was added. After 1 h stirring, the resulting gel beads were stored in 0.5 mol/L CaCl$_2$ solution for 1 h at room temperature. Na-alginate reacts with CaCl$_2$ to form beads and cross-linked CACH formed. During the process, the soluble sodium alginate converted into water insoluble calcium alginate beads. The resultant beads were stored after washing with de-ionized water to remove CaCl$_2$ from the bead surfaces. The beads collected by filtration and the excess water adsorbed on filter paper. The beads then dried at room temperature for 48 h. The loss of weight after drying was 98% and accompanied by an average diameter reduction of alginate beads from 3.17 ± 0.07 to 0.58 ± 0.34 mm. Volume decreased 97 ± 1.72 percentage after dried for 48 h at room temperature. The advantages of using dry alginate beads instead of the gel type are ease of handling and storage, greater precision during weighing and greater stiffness. In addition, synthesis steps by step shown in Figure 1.

Carl Zeiss 300VP SEM (Scanning electron microscope) (Figure 3) and PERKIN ELMER SPECTRUM TWO model FTIR-ATR Instrument (Fourier Transform Infrared Spectrophotometer) (Figure 4) were used for characterization of CACH.

![CACH synthesis](image)

**Figure 1.** CACH synthesis.

When 4 °C came 1-hour polymerization time is important to produce hydrogel of chitosan-alginate. Therefore, it is important to add Chitosan at that time. Before drying processes CACH particles shown in Figure 2.
As seen in the SEM images (Figure 3.) Chitosan particles successfully stuck in the calcium alginate hydrogels. In additions, when adsorption processes started, solution can enter CACH and interact with both hydrogel’s wall and chitosan particles.

FT-IR analysis shows at 2872.8 cm\(^{-1}\), Chitosan has a –NH\(^2\), NH\(^+\) and N-H bounds. However, inside of the CACH particle has not. Both particle has the other bounds (C=O, C-OH, C-O, C-O-C).

After synthesis hydrogel CACH particles was used as an adsorbent for adsorption of Sr (II) ions from aqueous solutions. To determine suitable conditions for adsorption, the effect of pH, contact time, concentration of Sr (II) ions, dose of CACH and finally the temperature effect experiments were investigated, respectively.
3. RESULTS AND DISCUSSION

Alginate is a natural polymer that may convert into hydrogels via cross-linking with divalent calcium ions. It has been preferred over other materials because of its advantages including biodegradability, hydrophilicity, presence of carboxylic groups and natural origin. The average size of gel particles estimated from SEM image was in the region of 0.4–0.93mm. Pores with different size observed. These pores are very suitable places for the adsorption of Sr (II) ions. The calcium alginate beads are stable in terms of their morphology after Sr (II) adsorption.

The Sr (II) uptake reached a maximum at pH 8.0 then it decreased. Therefore, pH 8.0 selected for further experiments. The initial concentration provides an important driving force to overcome all mass transfer resistance of Sr (II) between the aqueous and solid phases. The adsorption of Sr (II) carried out at different initial concentrations ranging from 15 to 100mg/L at pH 8.0 for CACH beads and the adsorption yield. The sorption has a maximum at 51% at the lowest Sr (II) concentration (15mg/L).

The effect of contact time on the adsorption of Sr (II) ions on CACH beads investigated over time intervals from 15 up to 180 min. Adsorption yield reaches maximum at about 120 min. In general, about 50% of the Sr (II) ion adsorption achieved within 120 min.

Adsorption data were processed according to different adsorption isotherms. The Freundlich (R²=0.96) and Langmuir (R²=0.91) isotherms are more compatible isotherms because their R² are closer to 1.

Figure 5. Adsorption of Sr (II) on CACH Isotherms a) Langmuir isotherm, b) Freundlich isotherm.

Thermodynamic studies showed that the adsorption of Sr (II) ions to CACH is exothermic. The greater the positive value of ΔG° with the increase of temperature means that larger energy is required for adsorption at high temperatures. (Table 1)

<table>
<thead>
<tr>
<th></th>
<th>ΔH (kJ mol⁻¹)</th>
<th>ΔS (kJ mol⁻¹ K⁻¹)</th>
<th>ΔG (kJ mol⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>298 K</td>
<td>303 K</td>
<td>308 K</td>
</tr>
<tr>
<td>(kJ mol⁻¹)</td>
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<tr>
<td>(kJ mol⁻¹ K⁻¹)</td>
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<tr>
<td></td>
<td>-0.04</td>
<td>-0.07</td>
<td>21.67</td>
</tr>
</tbody>
</table>
4. CONCLUSION

The removal of strontium ions from aquatic solutions was carried out using classical adsorption techniques. CACH beads were prepared and applied to remove strontium ions from the solution. The results of this study showed that chitosan-alginate composite hydrogel beads can be successfully used for strontium adsorption from aqueous solutions. Theoretical CACH’s Sr (II) adsorption capacity was calculated as \( Q = 17.3 \text{ mg/g} \) from Langmuir equation, under the optimum adsorption conditions (pH: 8, \( c = 25 \text{mg/L} \) Sr(II) concentration, \( t = 120 \text{ min} \), sorbent dosage: 4g/L and \( T = 298\text{K} \)).

The prepared chitosan alginate composite adsorbents exhibit good uptake capacities for Sr(II) ion. The CACH composites hydrogels can be readily separated from the solution adsorption by filter paper. CACH hold great promise as a potential adsorbent material and multifunctional platform for environmental applications. Therefore, the developed beads can be employed for the removal in continuous column operation mode. In this way, CACH has potential sorbent as water purification agent due to the different physical and chemical properties.

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REFERENCES


DETERMINATION OF $^{90}$Sr ACTIVITY CONCENTRATION IN TEA MATRIX AND UNCERTAINTY EVALUATION

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ABSTRACT
It requires many and different radioactivity measurements in order to radiation and environmental protection during the construction and operation of nuclear power plant. The determination of the strontium-90, which is highly radiotoxic fission product, in the environmental samples are also important in the routine environmental radiation monitoring. Many techniques such as co-precipitation, ion exchange etc. have been studied for the determination of radiostrontium in environmental samples. In this study, the method used for dissolution and radiochemical separation to determine strontium-90 activity concentration in tea matrix was investigated. Then, the strontium-90 activity concentrations were measured with ultra low-level liquid scintillation counter at the optimum conditions. The modified method was applied to proficiency test material containing contaminated Turkish black tea samples. Uncertainties arising from counting statistics, background, weighing, half-life, chemical recovery, efficiency and homogeneity were calculated.

KEYWORDS: Strontium-90, acid leaching, proficiency test, tea, uncertainty

1. INTRODUCTION
Most common methods used for radioactivity measurements such as gamma-ray spectrometry, alpha-particle spectrometry, liquid scintillation counting and gross alpha/beta-counting in Turkey are performed at the Department of Radiation and Accelerator Technologies, Turkish Atomic Energy Authority (which is a government institution). Also, there are many university and institute laboratories in Turkey currently measuring radioactivity or setting up new measurement systems. Radiation Metrology Division of the TAEA, the European Commission European Association of National Metrology Institutes (EC EURAMET) delegate on ionizing radiation metrology, is responsible for organizing a Proficiency Test (PT) to demonstrate the quantitative measurement capabilities of their laboratories on the identification of natural radionuclides in some matrices and to ensure the reliability of their measurement and analysis results (Karam et al., 2012; Şahin et al., 2016). The first PT organized by TAEA was performed on radiation measurement ($^{226}$Ra, $^{232}$Th and $^{40}$K) in soil matrices with twelve participating laboratories in 2013 (Şahin et al., 2016). The second proficiency test organized by TAEA was to determine radioactivity activities of $^{137}$Cs, $^{40}$K and $^{90}$Sr in contaminated Turkish black tea powder samples. Two set samples were sent to Joint Research Center-Geel (JRC-Geel) and Physikalisch-Technische Bundesanstalt (PTB-Germany) for the determination of the reference activity values of radionuclides in conjunction with TAEK. (Yeltepe et al., 2018).

The radioactivity measurement capabilities of laboratories in Turkey will be very important in near future because of an ongoing nuclear power plant program in Turkey. That’s why, the
measurement capabilities of radiostrontium in different matrices in national laboratories of Turkey should be enhanced especially for environmental contamination monitoring purposes.

In this study, the method used for determining the activity concentration value of $^{90}$Sr in tea matrix was investigated and then, the reference value of the second PT material prepared by TAEA was determined with the mentioned modified method. For a complete uncertainty budget, all uncertainty contributions to the calculation of $^{90}$Sr activity from the beginning of the experiment to the end of calculations such as counting statistics, weighing, decay correction, chemical recovery, efficiency and homogeneity were taken into account.

2. MATERIAL AND METHOD

2.1. Materials: A $^{90}$Sr/$^{90}$Y (38.60 ± 0.40 kBq g$^{-1}$) and tritium ($^3$H) (35.10 ± 0.80 kBq g$^{-1}$) certified reference solutions were also purchased from Physikalisch-Technische Bundesanstalt. Sr extraction chromatic resin columns were supplied by Eichrom Technologies, Inc. Teflon coated low diffusion polyethylene vials and Ultima Gold (UG) scintillation cocktail obtained from Perkin Elmer were used. All reagents were analytical reagent grade.

2.2. Samples: The details for the preparation and the homogeneity test concerning the PT material were reported in our previous studies (Yeltepe et al., 2018). The quench set was prepared by $^3$H standard reference solution, “15 mL UG plus 1 mL deionized water” cocktail and quencher agent (nitromethane). A commercial black tea which was sold from a market was used as blank sample. The commercially black tea sample (20 g) was also spiked with a stock standard solution with the activity of 67 Bq g$^{-1}$ prepared from $^{90}$Sr/$^{90}$Y certified reference solution for spike sample preparation.

2.3. Sample preparation: 20 g spike sample was put into a porcelain bowl. The sample was ashed in a microwave furnace for 16 h. A gradual heating program up to 600 ºC inside the microwave furnace was used to destroy organic matter. Then, sample was completely dissolved in 10 mL of 6.0 M HCl by heating and stirring up to 225 ºC and evaporated to dryness. By using 10 mL of conc. HNO$_3$ solution, sample was again dissolved and evaporated. This procedure was repeated three times. After 30 mL of 6.0 M HNO$_3$ was added, the sample was immediately centrifuged to remove the undissolved part from the solution. Thus, spike sample was ready for the radiochemical separation.

2.4. Radiochemical separation: A Sr-resin column was conditioned with 15 mL of 8.0 M HNO$_3$. The dissolved spike sample solution was loaded to the column. 15 mL of 3.0 M HNO$_3$ was added to the column to take $^{90}$Y radionuclide from the column. This eluate solution was discarded. And then, the column was washed with 20 mL of 0.05 M HNO$_3$ solution and eluate containing Sr-90 radionuclide was collected in a beaker. The collected eluate was evaporated up to nearly dryness. Finally, the residue taken up by 1 mL water was put into a scintillation vial containing 15 mL of UG scintillation cocktail for LSC measurement.

The same sample preparation and radiochemical separation procedure was applied for all samples. All samples were measured 3 times by LSC.

2.5. Instrument: Wallac 1220 Quantulus low background liquid scintillation counter (Turku, Finland) with an external standard of $^{152}$Eu was used for measuring count rate and external spectral quench parameter [SQP (E)]. Analytical results were evaluated by using WinQ and
Easy View software. Efficiency of radionuclide was determined with CIEMAT/NIST \(^3\)H efficiency tracing method (Malonda et al., 1982; Malonda et al., 1985). Milestone microwave ash furnace was used.

3. RESULTS AND DISCUSSION

The method used for assigning the \(^{90}\)Sr activity results in the contaminated tea matrix was developed. The developed method for \(^{90}\)Sr in the tea matrix was modified from the methods used for different matrices before (Aslan and Ozcayan, 2017; Aslan et al., 2015; Aslan et al., 2016; Altzitzoglou, 2016). The method is based on the digestion of the sample, the separation of \(^{90}\)Sr by column chromatography and the subsequent measurement of the activity by LSC. This sequential method for \(^{90}\)Sr was applied to the assessment activity concentration value of \(^{90}\)Sr in the second PT material prepared from the contaminated Turkish black tea powder including \(^{137}\)Cs, \(^{40}\)K and \(^{90}\)Sr.

In this study, the acid leaching procedure was the most important step of this modified method. Because each matrix has its own different complex and content. In the study, tea matrix was dissolved by using different acid concentrations and amounts in the dissolving step. Also, some modifications were made at the amounts of acid solutions used for conditioning and rinsing purposes in the radiochemical separation procedure according to the Eichrom method (ACW17VBS-2006).

Firstly, the prepared \(^3\)H quench set was measured in the LSC instrument at the beginning of this work. Then, the quench curve of tritium was plotted and fitted to a polynomial equation. Later, four spike samples at different activity concentrations were prepared. All samples were measured immediately using a Liquid scintillation counter with 1–600 window channels, 60 min counting time and three replicates. After measurement of \(^{90}\)Sr in tea matrix, the tritium efficiency values were calculated by using quench polynomial equation with spectral quench parameter values of \(^{90}\)Sr and then, the efficiency of \(^{90}\)Sr was calculated by using the following polynomial equations obtained from CIEMAT/NIST program.

The accuracy and precision of the method was evaluated by LSC measurements of prepared four spike samples at different activity concentrations (Fig. 1). The mean radiochemical recovery value was calculated as 78 %. In the experimental set, no outliers data were observed and the experimental relative standard deviation value of the mean RCV (7.28 %) was acceptable (10< %) (Aslan et. al, 2015).
Fig. 1. Mean recovery value of $^{90}$Sr spiked tea samples for the method (counting time= 60 min., 1-600 channels, 3 replicates, $\varepsilon_{\text{Sr-90}}$=99 %)

Two black samples identical to the spike material for the determination of background counting rate and detection limit were prepared and measured in LSC. The detection limit of the measured for $^{90}$Sr was calculated from the formula (y# equation) reported by ISO11929:2010 (Felice et al., 2017). The mean detection limit was found to be 1.1 Bq kg$^{-1}$ with 300 min. counting time and sample mass of 20 g.

The average activity of $^{90}$Sr in the second PT material was measured as 155 ± 16 Bq kg$^{-1}$ dry matter (k=2) by liquid scintillation efficiency tracing (CIEMAT/NIST) method by analyzing seven selected samples with three replicates for each sample. JRC-Geel took part in determining the activity values of $^{90}$Sr in PT material in conjunction with TAEA confirmed this value with the CIEMAT/ NIST method (Yeltepe et al., 2018). The measured $^{90}$Sr activity concentration values were given in Table 1.

Uncertainties come from counting statistics including background, weighing, counting efficiency, decay correction, chemical recovery, quenching were the most important components in the uncertainty budget for the activity concentration calculation of $^{90}$Sr activity from the beginning of the experiment to the end of calculations. Some uncertainty components such as wall effect, impurities, dead time correction, cocktails stability, kB value and half-life time were omitted to the final uncertainty budget with regard to previous studies (Yeltepe et al., 2018; Aslan et al., 2015; Altzitzoglou 2016; Kahraman et al., 2015). In this study, the main uncertainty contributions on the activity concentration for $^{90}$Sr were mostly coming from the uncertainty of the counting statics (1.24 %) and the chemical recovery (4.9 2 %). The reason of the uncertainty of the radiochemical recovery was due to standard deviation.
Table 1 Mean activity concentration results of $^{90}$Sr in the second PT material. All uncertainties are combined uncertainties at the 1σ level (k = 1) (counting time= 60 min., 1-600 channels, 3 replicates, RCV % = 78 %, $\varepsilon_{Sr-90}$=99 %)

<table>
<thead>
<tr>
<th>PT sample</th>
<th>Mean activity concentration with uncertainty (k = 1) (Bq kg$^{-1}$)</th>
<th>Mean activity concentration (Bq kg$^{-1}$)</th>
<th>Relative standard deviation, %</th>
<th>U activity (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PT_1</td>
<td>169.29 ± 8.74</td>
<td>155</td>
<td>5.16</td>
<td>8</td>
</tr>
<tr>
<td>PT_2</td>
<td>158.20 ± 8.16</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PT_3</td>
<td>147.91 ± 7.63</td>
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</tr>
<tr>
<td>PT_4</td>
<td>150.67 ± 7.77</td>
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</tr>
<tr>
<td>PT_5</td>
<td>151.02 ± 7.79</td>
<td></td>
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</tr>
<tr>
<td>PT_6</td>
<td>145.62 ± 7.51</td>
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<td></td>
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</tr>
<tr>
<td>PT_7</td>
<td>159.44 ± 8.23</td>
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</tbody>
</table>

4. CONCLUSION

Principally, sample preparation and radiochemical separation methods of matrices containing $^{90}$Sr radionuclide are the same but the amount and combination of acids and reagents used vary according to the matrix content. In this work, the optimum sample preparation method and measurement conditions of tea matrix containing $^{90}$Sr were determined. The $^{90}$Sr activity concentration used as reference values in the contaminated black tea for proficiency test sample was determined by using the modified dissolution and radiochemical separation procedures. Thus, the measurement capabilities of radiostrontium in different matrices in national laboratories of Turkey is increased by the time. When Turkey’s nuclear energy program is taken into the consideration, laboratories should be enhanced the comparability and reliability of radioactivity measurements for routine environmental radiation monitoring.

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5. REFERENCES


PLUTONIUM AND AMERICIUM IN THE DEEP BLACK SEA BOTTOM SEDIMENTS

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ABSTRACT

The new data on $^{239+240}$Pu, $^{238}$Pu, $^{241}$Pu, $^{241}$Am vertical distribution in the deep western Black Sea sediments was analyzed. The sediment dating by means of $^{238}$Pu/$^{239+240}$Pu activity ratio vertical distribution obtained suggested the upper 3 cm layer of sediments to be formed during 27 years after the Chernobyl accident and the deeper layers to be formed earlier in pre-Chernobyl time. The whole data set on profiles of radionuclides studied was shown to reflect the history of input into the basin and elimination of these radionuclides from the water column. The $^{241}$Am inventory after-deposition enhancement due to $^{241}$Pu decay was estimated by 74% in pre-Chernobyl sediment layer and the general contribution of $^{241}$Pu support in the total $^{241}$Am inventory in situ enhancement was estimated by 49%.

KEYWORDS: Black Sea; plutonium; americium; sediments; radionuclide inventory

1. INTRODUCTION

The world’s largest marine anoxic basin of the Black Sea is known to be affected by two major sources of anthropogenic radioactivity quite separated in time: the global radioactive fallout after nuclear weapon tests in open environments (peaked in early 1960s) and atmospheric fallout caused by accidental release from the Chernobyl nuclear power plant on 26 April 1986 followed by riverine discharge of significant quantities of radioactivity during early years after the accident (Polikarpov et al., 2008). Both mentioned sources contained a wide range of fission and activation products including plutonium isotopes (Polikarpov et al., 2008), which may be considered as one of the most informative tracers for the dating of seabed sediments (Gulin et al., 2002). The sediment dating by means of plutonium in the Black Sea is based on its isotopic composition: the $^{238}$Pu/$^{239+240}$Pu activity ratio was reported to differ over an order of magnitude – 0.036 for global fallout at the Black Sea latitude in 1971 and 0.45–0.50 for Chernobyl release in 1986 (Polikarpov et al., 2008). Besides the $\alpha$-emitting isotopes ($^{238,239,240}$Pu) both plutonium sources mentioned contained low-energy $\beta$-emitting $^{241}$Pu isotope being the parent radionuclide for $^{241}$Am and causing the $^{241}$Am increment in the environment (Livingston et al., 1987; Polikarpov et al., 2008). The $^{241}$Am isotope is known to have not been produced in any noticeable quantities directly in nuclear weapon explosions and to have been continuously produced in the environment from the $^{241}$Pu decay (Livingston et al., 1987). In contrast to global fallout for Chernobyl accidental release the presence of $^{241}$Am was reported, and the level of $^{241}$Am activity in Chernobyl fallout was shown to represent from 6.1 to 11.7% of $^{239+240}$Pu values obtained (Polikarpov et al., 2008).

The actual study aimed at analysis of $^{239+240}$Pu, $^{238}$Pu, $^{241}$Pu and $^{241}$Am vertical distribution in sediments from the western Black Sea deep area, dating of the sediments and estimation of $^{241}$Am after-deposition enhancement in different sediment layers due to $^{241}$Pu decay.
2. MATERIAL AND METHOD

Sediment samples for the actual study were collected with multiple corer equipment during 33rd international scientific cruise of RV Maria S. Merian in 2013 on the main station of the cruise (43°31.82′N–32°28.09′E, with water depth 2030 m). The core was sliced immediately after recovery using the rotary designed extruder into layers of 0.25 cm thickness within 0-3 cm sediment depth interval and 0.5 cm thickness down to 10 cm depth of the core.

Plutonium and americium isotopes were determined in the same samples by unified multistage radiochemical technique (Polikarpov et al., 2008). All samples were measured using EG&G Ortec Octete alpha-spectrometry equipment. $^{241}$Pu values were obtained using LKB Wallac Quantulus 2110 spectrometer. The sediment dating technique was based on geochronological reconstruction of plutonium contamination of the basin (Gulin et al., 2002). The part of $^{241}$Am gain occurred directly in bottom sediments after its deposition from water column (further called ‘in situ gain’) was estimated by equation (1) resulted from solving the system of equations describing $^{241}$Pu-$^{241}$Am parent-daughter decay and accumulation.

$$A_{Am}(t) = A_{0Pu} \cdot \frac{\lambda_{Am}}{\lambda_{Pu}} \cdot (e^{-\lambda_{Pu} t} - e^{-\lambda_{Am} t})$$

Values in equation (1) are: $A_{Am}(t)$ – $^{241}$Am in situ gain in each sediment layer during the period (t) from the layer formation year to the time of the core obtaining – 2013; $A_{0Pu}$ – activity concentration of $^{241}$Pu in each layer decay-corrected to the time of this layer formation derived from dating; $\lambda_{Pu}$ and $\lambda_{Am}$ are decay constants for $^{241}$Pu and $^{241}$Am respectively.

3. RESULTS AND DISCUSSION

Two well distinguishable $^{239+240}$Pu concentration peaks were obtained: the smaller one with 18.28 ± 1.91 Bq kg$^{-1}$ value was found in 2.75-3.00 cm sediment layer and lower in 4.0-4.5 cm layer $^{239+240}$Pu level was found to be the highest – 29.45 ± 3.79 Bq kg$^{-1}$. The $^{238}$Pu/$^{239+240}$Pu activity ratio in the 2.75-3.00 cm sediment layer was 0.34 ± 0.06. The last value suggests Chernobyl fallout to be the main cause of plutonium peak concentration in this sediment depth with up to 85 % (70% is the most mathematically expected value) contribution into plutonium origin in the layer. The 10 times lower $^{238}$Pu/$^{239+240}$Pu activity ratio (0.035 ± 0.009) in the 4.0-4.5 cm sediment layer indicates the global fallout to be the absolute dominant source of plutonium in this sediment depth. The $^{238}$Pu/$^{239+240}$Pu activity ratio in all sediments analyzed below the 3.00 cm depth did not exceed the value of 0.035 ± 0.010. Such $^{238}$Pu/$^{239+240}$Pu activity ratio vertical distribution suggests all layers deeper than 3.00 cm to be formed before 1986 as well as plutonium profile at these depths to reflect the pre-Chernobyl plutonium contamination history. So analyzing of the profiles obtained allows dating of 2.75-3.00 cm layer as 1986 and 4.0-4.5 cm – as 1963 (as maximum global radioactive fallout related) and on this basis one may proceed with converting proportionally the uncompacted sediment depth scale to the scale of time.

$^{241}$Pu activity concentration in Chernobyl peak was 353.9 ± 99.1 Bq kg$^{-1}$ (decay-corrected to 1986) and in global fallout peak – 247.5 ± 69.3 Bq kg$^{-1}$ (decay-corrected to 1963). The whole $^{241}$Pu profile was reconstructed by estimation of its concentration in each layer based on the isotopic composition of plutonium in all layers except ‘Chernobyl’ one presumed to be the
same and to be equal to the ‘global fallout’ peak. All values in this profile were decay-corrected to the data of layer formation according to the dating obtained.

The $^{241}$Am profile obtained is presented on Fig. 1. Two peak concentration levels of $^{241}$Am were obtained at the same sediment depths as for plutonium. $^{241}$Am activity concentration in the layer dated as ‘Chernobyl’ was 4.7 times higher than in ‘global fallout’ one. The percentage of estimated $^{241}$Am in situ gain from $^{241}$Pu decay in each layer where it was possible to calculate is presented on Fig. 1 as well.

![Fig. 1 - Vertical distribution of $^{241}$Am and estimation of its in situ gained fraction from $^{241}$Pu decay (absolute values – bars, relative enhancement – percentage numbers) in sediments from the deep western Black Sea](image)

The decrease of $^{241}$Am in situ gained fraction from values close to 100% in layers attributed to early years of man-made transuranium elements history in nature to units of percent in ‘post-Chernobyl’ sediment layers was obtained. The closeness of in situ gained $^{241}$Am fraction in the downmost layers to 100% is in good agreement with the fact that americium almost hadn’t been produced at all directly in nuclear explosions during weapon tests (Livingston et al., 1987), and from this point of view almost only plutonium including isotope $^{241}$Pu had been eliminated from the Black Sea water column at that time. The $^{241}$Am content eliminated from water column in 1950s can be considered to be negligible. The lack of $^{241}$Am during the 1950s in turn enhanced by the weak $^{241}$Pu content and yield in first fission bomb testing ($^{241}$Pu/$^{239+240}$Pu activity ratio – 0.75-2.5) in contrast to later thermonuclear events (Irlweck and Hrnecek, 1999). The further decrease of $^{241}$Am in situ gained fraction in upper ‘pre-Chernobyl’ layers reflects the increasing of americium sedimentation flux from the water column where the same process of its accumulation from $^{241}$Pu decay occurred. And this way the $^{241}$Am concentration in water must have been increased causing intensification of its elimination from the water column, especially taking into account that americium shows the more particle reactive behavior in marine environments than even plutonium. The further trend of $^{241}$Am in situ gain fraction decreasing in ‘post-Chernobyl’ layers amongst all mentioned points reflects reducing of the age of these layers to the top of the core and so reducing the time of in situ gaining process.

The fact of primary absence of $^{241}$Am in global fallouts is additionally reflected by its inventory values and the in situ gained fraction of these inventories. So the pre-Chernobyl inventory of $^{241}$Am was estimated to be gained in situ in sediments by 74% (7.3 Bq m$^{-2}$), and the absolute value of this inventory (9.9 Bq m$^{-2}$) is additionally resulted from the long period of the related layers existing (from thirty to sixty years old). The opposite pattern was obtained in the upper 3 cm of sediments where only 17% (1.2 of 7.4 Bq m$^{-2}$) of $^{241}$Am
inventory was estimated to be in situ gained from $^{241}\text{Pu}$ decay. The general contribution of $^{241}\text{Pu}$ support in the total $^{241}\text{Am}$ inventory in situ enhancement was estimated by 49%.

4. CONCLUSION

The alpha-emitting isotopes of plutonium and americium vertical distribution analysis in the deep Black Sea sediments was shown to reflect the history of these radionuclides input to the basin. The sediment dating suggested the upper 3 cm layer of sediments to be formed during 27 years after the Chernobyl accident and the deeper layers to be formed earlier in pre-Chernobyl time. The $^{241}\text{Am}$ inventory enhancement due to $^{241}\text{Pu}$ decay was estimated by 74% in pre-Chernobyl layers and the general contribution of $^{241}\text{Pu}$ support in the total $^{241}\text{Am}$ inventory in situ enhancement was estimated by 49%.

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5. REFERENCES


STUDY OF NATURAL RADIOACTIVITY AND ITS ASSOCIATED RADIOLOGICAL HAZARDS IN CERAMIC SAMPLES MANUFACTURED IN ALGERIA

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ABSTRACT

Ceramics are widely used in Algerian building constructions, they are considered as a source of hazardous contamination because of their naturally occurring radionuclides content. In order to estimate the harmful effects of ionizing radiations emitted by this material, broad investigations of natural radioactivity levels must be established. In this study, four ceramic samples manufactured in the east of Algeria were analyzed using a high-resolution HPGe semiconductor detector. Activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K were found in the ranges $(18.94±0.58 – 25.75±0.61)$, $(39.92±1.38 – 48.99±1.63)$ and $(418.86±10.54 – 644.39±10.87)$ Bq.kg$^{-1}$, respectively. These results are above the worldwide average ones for $^{40}$K and $^{232}$Th and below the recommended value for $^{226}$Ra. Radium equivalent activity ($Ra_{eq}$) was calculated and compared to the worldwide average value.

KEYWORDS: Ceramics; Radionuclides; Activity concentrations; Radium equivalent.

1. INTRODUCTION

Construction materials are commonly derived from soil and rocks. Therefore, they contain primordial radionuclides, such as $^{238}$U, $^{232}$Th and $^{235}$U series besides $^{40}$K. The segment of $^{228}$U decay chain starting from $^{226}$Ra is the most important from the radiological point of view. Hence, radium is often made as a reference instead of uranium (Sieu et al. 2011). These terrestrial radionuclides contribute to the major part of the natural radioactivity dose rate; they pose external exposure risks caused by their gamma-ray emissions and internal ones due to the inhalation of the radioactive inert gas radon ($^{222}$Rn) and its short-lived decay products (Solak et al. 2012). Ceramics are widely used as finishing materials in Algerian building constructions, they are considered as a source of hazardous contamination because of their naturally occurring radionuclides content. The presence of such primordial radionuclides in ceramics is due to their natural composition of clay, feldspar, silica, talc kaolin minerals and zirconium silicates (ZrSiO4). The main objective of this research work is to examine the activity concentrations of primordial radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K in four Algerian ceramic samples that are commonly used in dwellings. The measurements are realized by means of a high-resolution coaxial hyper-pure germanium detector. In the aim to assess the potential radiological risks related to the use of such material on inhabitant health, radium equivalent activity ($Ra_{eq}$) was calculated based
on the activity concentrations of the terrestrial radionuclides $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$. A comparison of the obtained values in this study with the worldwide average ones was made.

2. MATERIAL AND METHOD

2.1. Samples Collection and Preparation

Four ceramic samples were collected from ceramic dealers and construction sites, these samples are manufactured in the east of Algeria: C1 from Constantine, C2 from Batna, C3 from Béjaia and C4 from Bordj Bou Arreridj. They were crushed and milled into a fine powder, and then they were dried in an oven under a temperature of 110 °C for at least 24 hours in a way to remove the moisturizing content and obtain a constant weight. The samples were conditioned in thin Plexiglas boxes to avoid the self-absorption of low energy gamma. These boxes were carefully sealed for a period of four weeks to ensure the secular equilibrium.

2.2. Experimental Method for Gamma Spectroscopy

Sample measurements were carried out with a high-resolution HPGe semiconductor detector surrounded by 11.4-cm-thick graded lead shield (Canberra 7477 Series Lead Shield) and liners of 1.5 mm of copper and 3mm of tin to reduce X-ray fluorescence from lead shield. The gamma-energy photo-peak resolution of the gamma spectrometry system is 0.86 keV at 122 keV for $^{57}\text{Co}$ and 1.85 keV at 1332.5 for $^{60}\text{Co}$. The detector efficiency calibration was performed using a liquid source of $^{152}\text{Eu}$ mixed with water matrix. The detector efficiency values were also calculated by Monte Carlo simulation using MCNP5 code (Azbouche et al., 2015) taking into account the coincidence summing effects on efficiency calibration. Figure 1 shows the fitted efficiency curve as a function of gamma rays energy. The Genie 2000 software packages were used for the acquisition and treatment of the collected data. Each sample was measured for at least 24 hours to obtain the $\gamma$-spectrum with good statistics.

![FIGURE 1. Efficiency curve as a function of energies.](image-url)
2.3. Activity concentrations determination

Specific Activities of the radionuclides in the ceramic samples were determined using relation 1, taking into account the net peak area $N_{E_j}$ of a photopeak at energy $E_j$, the absolute full energy peak detection efficiency $\varepsilon_{E_j}$, the gamma-ray emission probability $P_{E_j}$, the counting time $t_c$ (s) and the mass of the measured sample $m$ (kg).

$$A_{E_j}(Bq/kg) = \frac{N_{E_j}}{\varepsilon_{E_j} \times P_{E_j} \times t_c \times m}$$

(1)

2.4. Radium equivalent activity ($Ra_{eq}$)

The distributions of $^{226}$Ra, $^{232}$Th and $^{40}$K are not uniform in the samples. Hence, Radium equivalent ($Ra_{eq}$) has been introduced to estimate the actual activity level of these radionuclides and to assess the radiation hazards associated with their presence in building materials. It is defined by equation 2 (Beretka and Matthew 1985). The base estimation of this formula is that 1 Bq/kg of $^{226}$Ra, 0.7 Bq/kg of $^{232}$Th and 13 Bq/kg of $^{40}$K produce the same $\gamma$-ray dose rate (Krisiuk et al. 1971).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K$$

(2)

3. RESULTS AND DISCUSSION

The activity concentrations of primordial radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K in the ceramic samples are shown in Figure 2. They are in the ranges (18.94±0.58 – 25.75±0.61), (39.92±1.38 – 48.99±1.63) and (418.86±10.54 – 644.39±10.87 Bq/kg, with mean specific activities of 23.04±0.62, 44.15±1.53 and 547.75±10.72 Bq/kg, respectively. The results, for $^{226}$Ra, are below the worldwide average one which is 40 Bq.kg$^{-1}$. For $^{232}$Th and K, they are above the recommended values of 40 and 400 Bq.kg$^{-1}$ respectively, as it is reported in the European recommendations based on a literature study (European Commission 1999; Mustonen et al. 1999).

FIGURE 2. Activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K for the investigated samples.
It can be clearly noticed that the sample C2 contains the highest activity concentrations of all radionuclides. While, sample C3 contains the lowest specific activities of $^{226}$Ra and $^{232}$Th. Nevertheless, sample C1 contains the lowest activity concentration of $^{40}$K. The Ra$_{eq}$ calculated values range from 115.97 Bq/kg to 145.43 Bq/kg, with an average value of 128.37 Bq/kg which is below the permissible limit of 370 Bq.kg$^{-1}$ (Beretka and Matthew 1985).

4. CONCLUSION

The natural radioactivity levels were measured in four ceramic samples manufactured in the east of Algeria. Measurements made with a high-resolution HPGe semi-conductor detector showed average activity concentrations of 23.04±0.62, 44.15±1.53 and 547.75±10.72 Bq/kg, for $^{226}$Ra, $^{232}$Th and $^{40}$K respectively. These values were below the recommended values for $^{226}$Ra, and above the worldwide values for $^{232}$Th and $^{40}$K. Radium equivalent activity (Ra$_{eq}$) was calculated for the studied samples and it have found to be lower than the worldwide average values, which indicate that these samples do not pose any radiation hazards.

5. REFERENCES


FEATURES OF THE PLUTONIUM RADIONUCLIDES DISTRIBUTION IN THE SALT LAKES OF THE CRIMEAN PENINSULA

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ABSTRACT

The levels of alpha-emitting radionuclides ²³⁸, ²³⁹+²⁴⁰Pu concentration activity in Crimean salt lakes from 4 groups of balneary and mud resources of the Crimea were studied. The ²³⁸, ²³⁹+²⁴⁰Pu in surface 0-5 cm layer in 10 lakes as well as their vertical distribution in 0-30 cm column of bottom sediment in four lakes were determined. The highest value of ²³⁹+²⁴⁰Pu in bottom sediment was in 10-15 cm layer in the Lake Sasyk-Sivash (2.1 Bq/kg) from the Yevpatoriya group. The average value of ²³⁹+²⁴⁰Pu in water for all groups of lakes was 6.5 and in the western Crimea – 11.5 mBq/m³, whereas in surface water of the Black Sea western part in near shore area – 0.5 mBq/m³. But the average ²³⁹+²⁴⁰Pu levels in the Crimean salt lake sediments in upper 0-5 cm layer to be 2-3 times lower than in the Black Sea coastal ecosystems. Salinity can be considered as one of the main factors influencing the distribution of plutonium in the lakes. Salinity ranged in the lake water within 50-330 ‰, except the Lake Kyzyl-Yar where present salinity was 4-7 ‰. The granulometric composition of sediments and features of the history of the primary and secondary contamination of plutonium radionuclides by air and water to salt lakes also played an important role for the ²³⁹+²⁴⁰Pu distribution in abiogenic components of the salt lake ecosystems. This is confirmed by the vertical profiles of plutonium radionuclides in the bottom sediments and the difference in the levels of plutonium accumulation in the sandy and silty bottom sediments of lakes, which differed by almost an order of magnitude.

KEYWORDS: salt lakes of Crimea, ²³⁸, ²³⁹+²⁴⁰Pu distribution, Black Sea, Chernobyl accident, global fallout

1. INTRODUCTION

Technogenic radionuclides began to input into the environment after humankind began using atomic technologies for both military and peaceful purposes. For the Crimean Peninsula, the main sources of man-made radioactive isotopes were global radioactive fallout after testing nuclear weapons in open environments, as well as radioactive fallout and input of radionuclides with the river runoff after the Chernobyl accident (Warner and Harrison, 1993; Polikarpov et al., 2008). Aquatic ecosystems play important role in migration of radionuclides. Lakes occupy a special place among aquatic ecosystems, because many of them are stagnant reservoirs and therefore they become a long-term depot for radionuclides. The lakes are widely used by people and at the same time they serve as radionuclide depositories. So study of distribution and inventory of man-made radioisotopes in lakes is important part of radioecology of water ecosystems.
The plutonium radioecology in the salt lakes of the Crimea is of special interest because they have unique resources (Oliferov and Timchenko, 2005). The water of the salt lakes is a valuable raw material and balneological resource as well as bottom sediment. Salt lakes are also a source of biological resources. Studies of salt lakes in the Crimea to determine $^{238,239,240}$Pu in the components of their ecosystems until recently, did not perform. At first in 2016 we studied the level of $^{239+240}$Pu and $^{238}$Pu in upper 0-5 cm layer of Crimean salt lake bottom sediments (Tereshchenko et al. 2018). The purpose of this investigation was to research the levels of man-made plutonium radioisotopes $^{239+240}$Pu and $^{238}$Pu in bottom sediments as well as in water in 10 large salt lakes of the Crimean Peninsula from 4 geographical (territorial) groups (Fig. 1.) including vertical distribution of $^{239+240}$Pu and $^{238}$Pu in bottom sediments in layer 0-30 cm in the lakes of each group and comparative analysis of plutonium radionuclide levels in lakes and adjacent Black Sea areas.

2. MATERIAL AND METHOD

We studied 10 Crimean salt lakes from 4 geographical groups (Fig. 1). Materials for investigation were taken during coastal land expeditions in 2016 – 2018.

![Fig. 1. Schematic map of 10 studied salt lakes location in Crimea from 4 geographical groups of Crimean lakes: 1 – Kyzyl-Yar, 2 – Sasyk-Sivash, 3 – Dzharylhach, 4 – Bakal, 5 – Krasnoye, 6 – Kiyat, 7 – Kirleut, 8 – Aktash, 9 – Chokrak, 10 – Tobechik, where (dashed line): Ye.G. – Yevpatoriya group, T.G. – Tarkhankut group, P.G. – Perekop group and K.G. – Kerch group; black star – sampling stations in the coastal zone of the seas.]

The $^{238, 239+240}$Pu determined in natural objects by a known method (Tereshchenko et al., 2018). The radioactive tracer $^{242}$Pu was added to the samples after they have cooled down. Then chemical decomposition of the samples was carried out to convert plutonium into a dissolved state. Further purification and separation of plutonium was carried out using ion exchange chromatography with an anion exchange resin (Tereshchenko et al., 2018a). Plutonium thin-layer preparations on a plate were measured on the alpha-spectrometric complex "EG & G ORTEC OCTETE PC" in the department of continental radioecology at the Institute of Plant and Animal Ecology in the Ural Branch of the Russian Academy of Sciences (Ekaterinburg) and in the department of radiation and chemical biology in the A. O. Kovalevsky Institute of Marine Biological Research (Sevastopol). Results of the $^{238, 239+240}$Pu concentration activity determining are presented as: mean ± standard deviation. The total relative error of determining the $^{239+240}$Pu activity concentration did not exceed 20%. However the Pu determining error was above 50% for the recent years water samples because of the low levels of its activity concentration in some samples of bottom sediments and sea water especially for $^{238}$Pu. In some samples the $^{238}$Pu activity concentration was below limit detection.
3. RESULTS AND DISCUSSION

The $^{238,239+240}$Pu activity concentration was measured in all 10 lakes during 2016-2018. The highest $^{239+240}$Pu activity concentration was observed in silt bottom sediments (0-5 cm) from lakes of different groups: the Lake Kyzyl-Yar – $426 \pm 27.3$, Dzharylhach – $440 \pm 24.4$ and Chokrak – $385 \pm 44.3$ mBq/kg (marine origin lake’s groups: Yevpatoriya, Tarkhankut and Kerch, respectively) and the lowest one – in three investigated lakes of the Perekop group (continental origin group of lakes) where level of $^{239+240}$Pu changed from $22 \pm 20.3$ to $67 \pm 35.7$ mBq/kg. Such a radioecological situation in the lakes of the Perekop group may be due to on the one hand the continental origin of the lakes and lack of connection these lakes with the sea waters in the modern period.

The vertical distribution profiles of these radionuclides in sediment cores with a depth of 30 cm were obtained in four lakes. The highest value was recorded in 10-15 cm sediment layer in the Lake Sasyk-Sivash (2.1 Bq/kg) from the Yevpatoriya group and the ratio of $^{238}$Pu/$^{239+240}$Pu indicates the global origin of plutonium at this depth. In different lakes, global fallout peaks occurred at depths of 10–20 cm. The $^{239+240}$Pu levels at these depths were 0.6–2.1 mBq/kg and their differences between the lakes were significantly less than in the surface layer. The vertical profiles of $^{238}$Pu, $^{239+240}$Pu in four lakes are shown in Fig. 2. The $^{238+239+240}$Pu inventory in upper 0-30 cm layer of bottom sediments in 4 investigated salt lakes changed from 86.5 to 196.2 Bq/m$^2$. It was estimated for 2016.

![Fig. 2. The vertical profile of $^{239+240}$Pu, $^{238}$Pu in bottom sediments from four lakes of studied balneological and mud resources group of the Crimean salt lakes.](image)

4. CONCLUSION

1. It was established that in the studied salt lakes of the Crimea, the activity concentration of $^{239+240}$Pu in the water was 0.8–16.5 mBq/m$^3$ and exceeds that in the Black Sea coastal waters by 13 times.

2. It was determined that the levels of $^{239+240}$Pu in the surface layer of bottom sediments of salt lakes varied within 0.024-0.516 Bq/kg, which was lower than in the coastal Black Sea regions (0.3-1.8 Bq/kg).
3. The levels of $^{239+240}$Pu in the sediments of lakes of marine origin (lakes of the Yevpatoriya, Tarkhankut and Kerch groups) were on average 5 times higher than those of lakes of continental origin (lakes of the Perekop group).

4. The data on the $^{239+240}$Pu, $^{238}$Pu vertical distribution and the ratio of activities $^{238}$Pu/$^{239+240}$Pu indicate the presence of an individual history and intensity of $^{238+239+240}$Pu input in different lakes and a large proportion of plutonium was global origin.

5. It was shown that concentration factor of $^{239+240}$Pu $C_d(239+240)$Pu for bottom sediments of lakes reached high values and was equaled $n \times 10^5$, but it was lower compared to $C_d(239+240)$Pu of the Black Sea bottom sediments ($n \times 10^6$).

6. The integrated deposition density of the total $^{238+239+240}$Pu in 0-30 cm layer in bottom sediments of four salt lakes ranged from 86.5 to 196.2 Bq/m$^2$ (for 2016).

7. The results of study indicated that the levels of plutonium radionuclides in bottom sediments differed both between the four resource groups of the Crimean lakes and within the groups. The depth of the layer of bottom sediments with anthropogenic plutonium radionuclides in different lakes was limited to 15-35 cm of sediment. In the Lake Chokrak, this layer extends deeper than 30 cm. Different conditions of the geographical location of lakes, their catchment basins, and biogeochemical characteristics of lake ecosystems influence the formation of levels and depths of plutonium radionuclides. This causes individual quantitative characteristics in each lake and in each group of balneological and mud resources of the Crimean lakes, which indicates the need for further research of lakes.

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5. REFERENCES


DOSE AND RISK ESTIMATION OF Cs-137 AND I-131 RELEASED FROM A HYPOTHETICAL ACCIDENT IN AKKUYU NUCLEAR POWER PLANT

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ABSTRACT
The construction of Akkuyu Nuclear Power Plant (NPP) was launched in 2018 and the plant is expected to be operative by the year 2023. Being situated in the Mediterranean coastline, Akkuyu NPP will be the first nuclear power generation facility in Turkey. The plant will have four Russian VVER-1200 type pressurized water reactors with a total installed capacity of 4800 MW. In this study, atmospheric dispersion and ground level deposition of Cs-137 and I-131 released from a possible accident in Akkuyu NPP was estimated using a Lagrangian particle dispersion model, FLEXPART, for different time periods representing relatively extreme meteorological conditions for Mersin. The source term used in simulations was assumed to be the same with that of the Chernobyl NPP accident that occurred in 1986. In addition, cumulative dose and risk values were calculated from FLEXPART output datasets considering potential exposure pathways such as inhalation, ground-shine exposure and cloud-shine exposure. The results were further analyzed with python codes and dose and risk maps were created for local and regional scales. The outputs were finally compared with past nuclear accidents and related risk studies in literature. According to results of the study, it was found that the vicinity of Mersin and Central Anatolia were simulated to be the most significantly affected areas from the accident under both scenario conditions. The northern and western parts and all coastlines of Turkey were simulated to be more contaminated in the simulations conducted under December 2009 conditions, whereas southern and western parts of Turkey and some parts of Middle East countries like Syria, Iraq and Lebanon were simulated to be comparatively more contaminated under the runs conducted for August 2010 period. Thus, it can be clearly stated that the meteorological conditions were among the most important parameters for the fate and transport of radioactivity that originated from such a catastrophic event.

KEYWORDS: nuclear power plant, dose estimation, risk assessment, atmospheric fate and transport modeling, Lagrangian particle dispersion

1. INTRODUCTION
The foundation of Akkuyu Nuclear Power Plant (NPP), which is the first nuclear power plant of Turkey, was laid in 2018. It was also announced that electricity generation in the plant would start in 2023. The environmental impacts of uncontrolled radionuclide releases from previous or potential future accidents have been the topic a number of research. These studies
primarily focused on the dispersion patterns of radioactive particles in the atmosphere and the associated fallout that creates deposition of radioactivity on the ground.

In this particular study, a mathematical model was developed in FLEXPART platform to estimate the atmospheric dispersion and ground level deposition of radionuclides to be released from a possible nuclear accident in Akkuyu NPP under different atmospheric conditions. The results obtained were converted into dose values considering various exposure pathways. All results were visualized and compared the past accidents and similar studies for the evaluation.

2. MATERIAL AND METHOD

A Lagrangian particle dispersion model, FLEXPART (Stohl et al., 2005), was used in this study to simulate dispersion of the radionuclides released from a potential nuclear accident in Akkuyu NPP. FLEXPART model utilizes ECMWF and GFS (NCEP) data as the meteorological input. For this particular study, a 0.5x0.5 degree resolution, 6-hourly NCEP reanalysis data were used in simulations. Based on climatic status of Mersin province, two different periods were determined for model simulations. The first period was selected to be December 2009, which is considered to have the highest precipitation. The second simulation period was selected to be August 2010, which represented the hottest and relatively driest period according to the long-term meteorological data from the nearest Mersin station, which approximately characterizes Akkuyu NPP site.

Two radionuclides (Cs-137 and I-131) with relatively long and short half-lives, respectively, were simulated in this study. Due to the relatively new design, there is no officially published source term information in the literature for VVER-1200 reactor. Therefore, the source terms estimated by Evangeliou et al. (2017) of Chernobyl, which is considered the worst nuclear power plant accident in the history, were used in this study.

Three different dose values were also estimated using dispersion and deposition results of the study considering cloudshine, groundshine and inhalation exposure pathways: contribution of Cs-137 on the effective dose for 1 year and 50 years, and 7-day thyroid dose from I-131. Ingestion pathway is typically neglected in literature because accurate evaluation of this term requires information on population and on food consumption, which is often difficult to obtain. As a result, ingestion pathway was also neglected in this study.

Dose coefficients were taken from a report prepared by a joint working group in Canada (Health Canada, 1999). The coefficients reported in Health Canada (1999) were taken from ICRP (1996) and Eckerman and Legett (1996) for internal and external exposures, respectively. Although radiation has different effects on children and adults, dose calculations were made only for adults in this study.
3. RESULTS AND DISCUSSION

Deposition Results

When the deposition results are analyzed, it can be seen that significant depositions were observed in western parts of Turkey, where population intensity is high, during the simulation conducted for December 2009 period. Almost, all places in western Turkey were contaminated with levels higher than 1 kBq/m² of Cs-137. These results also reveal the significant topography effect on depositions along the coastal regions. Depositions are relatively higher in the entire coastline of Turkey as well as some coastlines in other countries in the Mediterranean and the Adriatic Sea (Figure 1).

Simulation conducted for August 2010 period demonstrated that the atmospheric transport was relatively towards south direction. In this scenario, Cs-137 was predominantly deposited near Mersin and Antalya, in some parts of central Anatolia, western Syria and a portion of Egypt. Northern territories of Turkey was less influenced in comparison to southern territories. Since the amount of precipitation is almost zero in this season, higher deposition values were observed compared to Scenario 1. The contamination around Antalya occurred due to radionuclides transported via higher-level air movements in the 4th day. This situation demonstrated that vertical variations in release patterns and rising height of contaminant species are significant factors on the general distribution of contamination. (Figure 1).

Dose Estimation Results

Three different dose estimations were performed in this study. These included (i) a 7-day thyroid dose for I-131, (ii) a 1-year mid-term dose for Cs-137 and (iii) a 50-year lifetime dose for Cs-137. Dose estimations considered all possible pathways except ingestion. Ingestion pathway was neglected from all dose estimations due to difficulty in supplying the necessary data. As dose estimations normally incorporate the effect of all emitted radionuclides from the accident, measured doses are typically higher than what is reported in this study. It should be noted that the dose estimations performed herein only included the dose associated with I-131.
and Cs-137. Consequently, collective doses values simulated in this study were found to be lower than the expected range observed after the Chernobyl event.

The 7-day thyroid dose calculations covered the entire simulation period and the time window that gave the highest 7-day cumulative value were reported to be the thyroid dose. The calculation of thyroid dose caused by respiration of I-131 demonstrated considerable differences for both simulation periods. Maximum values were obtained between 06.12.2009 09:00 and 13.12.2009 12:00 and between 07.08.2010 18:00 – 14.08.2010 21:00 for the first and second scenarios, respectively. In the first scenario, dose values ranged between 1 to 10 mSv that were spatially distributed around Mersin, east of Antalya, west of Central Anatolia and the vicinity of the City of İzmir, which is the third most populated metropolitan area of Turkey, and western Syria. Relatively high values were detected in eastern sides of the source in the second scenario (Figure 2).

![Figure 2. Seven-day Thyroid dose caused by I-131 in Scenario1 and Contribution of Cs-137 on Effective Dose for 1 year in Scenario 2.](image)

The results of 1-year dose calculation of Cs-137 demonstrated that higher values were observed in northern and western territories of the simulation domain in 2009 whereas in southern and eastern portions in 2010 as was found in the 7-day thyroid estimations. However, it was revealed that contribution on 1-year collective dose of only Cs-137 release was calculated to be less than the 7-day inhalation dose of I-131. The reason of this difference was the total release amount of I-131. The source terms value of I-131 release was 15 times higher than of Cs-137, which was reflected in the total dose values calculated for the domain.

4. CONCLUSION

The simulations conducted in this study also highlight the fact that radiological contamination occurring from Akkuyu NPP is likely to influence other countries in the immediate vicinity. Therefore, it can be concluded that the construction and operation of a nuclear power plant is not only a local or national topic but also an issue of international scale. Thus, multi-national studies need to be conducted in nuclear risk assessment process.
The vertical mixing length and local meteorological conditions at the accident site are found to be extremely important in the overall dispersion of contamination. Such factors are quite influential on the fate and transport of radionuclides. Meteorological conditions define the lower and upper mixing boundaries in the atmosphere, which is also a function of seasonal timing of the accident. Therefore, vertical changes in release patterns should be analyzed extensively to cover all possible conditions that are likely to occur from a hypothetical accident.

The local morphology of the accident site and its vicinity are also influential in the results. These factors have direct influence on seasonal precipitation patterns. Power plants situated in coastal zones that are backed by steeply sloping mountain ranges are likely to receive more convective precipitation, a major removal mechanism for radioactivity. Thus, the effect of local topography and micro climatological conditions are also influential in the dispersion of contamination.

Last but not the least, simulation results are strongly influenced from the meteorological conditions. It is observed that meteorological data is a critical component of the simulation process and different meteorological data sets are likely to reveal different results even if all other parameters are kept constant. In this regard, simulations need also be conducted with different datasets such as ERA-INTERIM, ERA-5 and others.

REFERENCES


DETERMINING $^{90}$Sr/$^{137}$Cs RATIOS IN SOIL SAMPLES AT THE OBJECTS OF THE «EXPERIMENTAL FIELD» OF THE SEMIPALATINSK TEST SITE

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ABSTRACT

This paper provides results of researching $^{90}$Sr to $^{137}$Cs ratio values is soil of epicentral zones, as well as the territories, subjected to radioactive fallouts, within the «Experimental Field», designed for surface and atmospheric nuclear tests of various yields from 1949 till 1962. Radioactive contamination of the site area is characterized by fission products ($^{137}$Cs, $^{90}$Sr), neutron activation products ($^{152}$Eu, $^{154}$Eu, $^{60}$Co) and nuclear charge material ($^{241}$Am, $^{239+240}$Pu). The determining $^{137}$Cs activity is used cheap and fast gamma-spectrometric method, however classical methods of determining $^{90}$Sr activity are labor intensive and expensive. Alternatively, $^{90}$Sr activity can be assessed using $^{137}$Cs activity. According to the literature data, the $^{90}$Sr/$^{137}$Cs ratio value depends on the type of nuclear substance and it remains unchanged for the same test. For a $^{235}$U-based nuclear charge the $^{90}$Sr to $^{137}$Cs ratio equals to 0.88; for a $^{239}$Pu-based charge this ratio equals to 0.3. As the result of research the $^{90}$Sr/$^{137}$Cs ratio at the radioactive fallout plumes and in epicenter zones was found to range from 1.0 to 2.5 and from 0.4 to 4.8 respectively. The majority of experimentally obtained $^{90}$Sr/$^{137}$Cs ratio values are beyond the theoretical assessment interval. By now this issue remains issue. The main result of this work is the conclusion, each researched object at the «Experimental Filed» site should be characterized by its own $^{90}$Sr/$^{137}$Cs ratio, since the ratio values differ between different objects of the researched site depending on origin of radioactive contamination.

KEYWORDS: Semipalatinsk Test Site (STS), «Experimental Field» site, $^{90}$Sr/$^{137}$Cs ratio, soil

1. INTRODUCTION

It is known the soil is the main component of the natural environment that is subject to radioactive contamination during nuclear testing. All substances in soil, that pollute the ecosystem, including artificial radionuclides, accumulate and remain. Therefore, the study of the content and distribution of artificial radionuclides in the soils of the Semipalatinsk test site is a very important aspect of researches. The main source of surface radioactive contamination of soil at the STS is the «Experimental Field» site. 116 nuclear atmospheric and surface tests were conducted there. All tests at the «Experimental Field» were conducted at 5 technical sites as follows: P-1, P-2, P-3, P-5, P-7. Thirty surface nuclear tests conducted there had the yield ranged from 0.007 to 400 kT of TNT equivalent. These tests have caused radioactive contamination of soil cover both in the testing area of the «Experimental Field», and in the area of plumes, extended far beyond the STS. At the present time, radioactive contamination of the «Experimental Field» is caused by remaining fission material of the nuclear charge, neutron activation products and long-living fission fragments [i, ii].
Assessment of radiological situation at the radiation-hazardous objects of the «Experimental Field» testing site requires to know concentrations of such radionuclides as ⁹⁰Sr and ¹³⁷Cs in soil, having long half-life period, high migration ability and high yield in fission reactions.

The parent isotopes of these radionuclides are short-living elements, which mostly decay into ⁹⁰Sr and ¹³⁷Cs. ¹³⁷Cs activity is determined using fast and pretty cheap gamma-spectrometric method, while currently available methods of determining ⁹⁰Sr low activity concentrations are labor- and cost- ineffective. It is clear, that this method is not acceptable for mass analysis. As an alternative we can use a ratio, allowing to assess activity of one radionuclide, using known activity of another.

Here on the diagram you can also see ⁹⁰Sr to ¹³⁷Cs specific activity ratios in the first hour after the nuclear explosion for different fissionable materials: ²³⁵U and ²³⁹Pu. As you can see on the diagram, ⁹⁰Sr/¹³⁷Cs ratio for ²³⁵U and ²³⁹Pu is 0.9 and 0.3 respectively.

The most interesting here is researching the possibility of determining ⁹⁰Sr activity using known ¹³⁷Cs activity in soil of different objects of the «Experimental Field » using calculation method.

2. MATERIAL AND METHOD

Object of research. As the objects of research the epicentral zones of 5 technical sites as follows: P-1, P-2, P-3, P-5, P-7 and two radioactive fallout plumes that spread in the southwest and southeast directions of the «Experimental Field» were chosen.

Research methodology. The research methodology consisted of soil sampling, preparation of soil samples, determination of quantitative characteristics of radioactive contamination with ⁹⁰Sr and ¹³⁷Cs fission products, calculation of ⁹⁰Sr/¹³⁷Cs ratios in the epicentral zones at the «Experimental Field» site, statistical analysis and interpretation of results.

Sampling. Soil samples were collected using point method in the epicentral zones and along the axis of radioactive fallout plumes at the depth of 0–5 cm and the area of 10 cm². This depth was chosen on purpose – the maximum concentration of artificial radionuclides in arid zone is in top layers 0-5-10cm, although already more than 55 years have passed since the last surface test was conducted [iii].

Sampling preparation. Soil samples were dried at the temperature of 105 °C to absolutely dry state. Dried soil sample was sieved using the 1 mm mesh sieve. Coarse fraction with the size of >1 mm was not used in further studies. From the fraction of <1 mm about ~ 150 g subsample was taken using squaring method. After that the soil subsample was milled via PULVERISETTE 9 disk laboratory mill for 20 minutes at the rotation speed of 1000 rpm.

Radionuclide analyses. The activity concentration of of ¹³⁷Cs was determined using BE5030 gamma-spectrometer with semiconductor detector («Canberra», USA). The exposure time ranged between 1 and 2 hours, it was depending on activity of samples. Correctness of ¹³⁷Cs specific activity determination was controlled by means of periodic measurements of standards (OMASN, Russia). Specific activity of ⁹⁰Sr was determined by means of direct measurement with «Progress-BG» beta-spectrometer («Doza», Russia). The exposure time was 20 minutes. Correctness of ⁹⁰Sr specific activity determination was controlled by means of periodic measurements of ²²Na control calibration source. In case of specific activity
concentration of the radionuclide was below the detection limit (100 Bq/kg), the subsamples were prepared for radiochemical extraction of $^{90}$Sr. The $^{90}$Sr activity concentration was determined from the activity concentration of Y-90 by using a liquid scintillation beta-spectrometer «Tri-Carb 2900 TR» («PerkinElmer», USA). The exposure time was 2 hours.

Statistical analysis Only numerical values of these radionuclides used for calculation of the isotopic ratio of $^{90}$Sr/$^{137}$Cs. Also, statistical analysis of the ratios was carried out, namely: selection (n), the arithmetic mean (µ), the correlation coefficient (R), standard deviation, determination abnormally high values and their exclusion from the general dataset using the normalized deviations. Checking for compliance with normal distribution was performed using Pearson's test.

3. RESULTS AND DISCUSSION

The $^{90}$Sr/$^{137}$Cs ratios in the soil of epicentral zones. The fission products ratio was calculated for each epicenter separately, and also the arithmetic average (or geometric average) ratio – for the technical site as a whole.

On the technical site P-1 there is one epicentral zone. 10 soil samples were collected in the epicenter zone. The $^{90}$Sr/$^{137}$Cs ratio factor of 4.8 was obtained with a good correlation factor of 0.96. Calculated $^{90}$Sr/$^{137}$Cs ratio values ranged between 2.9 and 5.5. Mean root square deviation was 0.8.

On the technical site P-3 there are 2 epicentral zones. 3 soil samples were taken from each epicenter of this site. For P-3 technical site a $^{90}$Sr/$^{137}$Cs ratio factor of 1.0 was obtained at the correlation factor of 0.64. Calculated $^{90}$Sr/$^{137}$Cs ratio values ranged between 0.5 and 3.4, at that 3.4 is the outlier, after removal of that ratio values of fission products ranged between 0.5 and 2.1. Mean root square deviation was 0.4.

At site P-5 were registered 6 epicentral zones. Similar to P-3, 3 soil samples were taken from each epicenter. For P-5 technical site $^{90}$Sr/$^{137}$Cs ratio factor of 1.1 was found at the correlation factor of 0.95. Calculated $^{90}$Sr/$^{137}$Cs ratio values ranged between 0.2 and 2.1. Mean root square deviation was found to be 0.5.

$^{90}$Sr/$^{137}$Cs fission products ratio in the epicentral zones of technical sites P-2, P-7. These technical sites are considered together, because of their location peculiarities that they overlap one another. At P-2, P-7 sites 19 pronounced epicentral zones were registered. Similar to P-3 and P-5, 3 soil samples were taken from each epicenter. For P-2, P-7 sites $^{90}$Sr/$^{137}$Cs ratio factor of 0.8 was obtained with a good correlation factor of 0.89. Calculated $^{90}$Sr/$^{137}$Cs ratio values ranged between 0.4 and 5.1. After exclusion of outliers, calculated values ranged between 0.4 and 2.8. Mean root square deviation was 0.6.

The results of the researches at the epicenters of the «Experimental Field» site are shown in the Table 1.

<table>
<thead>
<tr>
<th>Technical site</th>
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<tbody>
<tr>
<td>Epicenter number</td>
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<tr>
<td>P-1</td>
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<td>P-3</td>
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<td>P-5</td>
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Table 3. The $^{90}$Sr/$^{137}$Cs ratios in the soil of epicenters of the «Experimental field» site
It should be noted, that $^{90}\text{Sr}/^{137}\text{Cs}$ ratio coefficients obtained at the researched technical sites significantly differ from one another. This fact speaks of the difference in sources of radioactive contamination. According to historical data, these sites were used for nuclear tests with high, moderate, low and super low yield. Also there were emergency nuclear tests. In addition, it is unknown exactly, what epicenter to what test belongs (by the date of testing).

The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in the soil of the plumes of the nuclear test. Plumes of fallout from nuclear tests are parts of the terrain, the surface radioactive contamination of which is formed by the deposition of radioactive particles on the surface of the earth, often in the form of a strip originating from the epicenter. As part of this work, two plumes are considered, located at the «Experimental Field» site.

The plume of the nuclear test conducted in 1953. This plume was formed as a result of the first thermonuclear test of August 12, 1953, for 400 yield in TNT equivalent. It extends hundreds km from the P-1 technical site’s explosion epicenter, but in this work is researching just part of this plume, from its epicenter to the boundary of the «Experimental Field» site, in the southwest. Its length is about 8.5 km. 25 soil samples were collected on the plume. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratio factor of 2.5 was found at the correlation factor of 0.80. Calculated $^{90}\text{Sr}/^{137}\text{Cs}$ ratio values ranged between 1.8 and 3.4. Mean root square deviation was found to be 0.8.

The plume of the nuclear test explosion of plutonium charge. This plume was formed as a result of the nuclear test explosion of plutonium charge. This plume is located in southeast directions of the «Experimental Field». Its length is about 2 km. 8 soil samples were collected on this plume. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratio equals to 1.3 at the correlation factor of 0.99. Calculated $^{90}\text{Sr}/^{137}\text{Cs}$ ratio values ranged between 1.0 and 2.9. Mean root square deviation was found to be 0.69.

Some of the plumes parts on technical sites P-2, P-7 were remediated (ploughed). As the result of that it was not possible to conduct a comparative analysis of ratio fission products.

The results of the researches on the plumes of radioactive fallout of the «Experimental Field» site are shown in the Table 2.

Table 4. The $^{90}\text{Sr}/^{137}\text{Cs}$ ratios in the soil of the plumes of radioactive fallout of the «Experimental field» site.
The plume  |  \(^{90}\text{Sr}/^{137}\text{Cs} \) ratio of epicenter
--- | ---
B-1 | 1.3
The part of the plume from the test of 12.08.1953 | 2.5

\(^{90}\text{Sr}/^{137}\text{Cs} \) ratios on plumes of radioactive fallout differ from each other. This fact suggests of the difference in nuclear charges, different height of the charges and explosion yields.

4. CONCLUSION

All these \(^{90}\text{Sr}/^{137}\text{Cs} \) ratios are within narrow range. It makes possible to assess activity of \(^{90}\text{Sr} \) by activity of \(^{137}\text{Cs} \) in soil researched objects of the «Experimental Field» site using own \(^{90}\text{Sr}/^{137}\text{Cs} \) ratio suitable for each object. Also, \(^{90}\text{Sr}/^{137}\text{Cs} \) ratios in soil of the «Experimental Field» site differ from \(^{90}\text{Sr}/^{137}\text{Cs} \) ratios in soil of background territories (\(^{90}\text{Sr}/^{137}\text{Cs} = 0.6 \) ). It enables us to determine boundaries between background territories and nuclear test sites. Ratios of fission products using can allow for optimizing radioecological researches by avoiding cost ineffective radiochemical analyses of \(^{90}\text{Sr} \) and using them only to proof the obtained results.

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