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VERTICAL DISTRIBUTION OF RADIONUCLIDES IN SOILS OF SEMIPALATINSK TEST SITE

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The article addresses the pattern of vertical distribution of the major long-lived man-made radionuclides ^{137}Cs, ^{241}Am, ^{90}Sr and $^{239+240}Pu$ in soils of the Semipalatinsk Test Site. Areas of different contamination with radionuclides are discussed – places of aboveground nuclear and fusion tests conducted at the 'Experimental Field' site, the fallout in the form of plumes within the Semipalatinsk Test Site, areas of radiological warfare agent tests at the '4A' site, areas of meadow ecosystems associated with radioactively contaminated water streams from test adits of the 'Degelen' testing site, conventionally 'background' areas of the test site, in which no nuclear or fusion tests were conducted. In the course of research, differences were revealed in the vertical distribution of radionuclides of interest in soils of the above areas. Differences are attributed to the pattern of how contamination with radionuclides is formed and to abiotic and biotic factors such as physical and chemical soil properties, moistening conditions, human activity and others. Based upon findings, recommendations were developed, aimed at the optimization of research into the vertical distribution of radionuclides in the soil cover of the former Semipalatinsk test Site. In particular, it was found that when undertaking such research, it was sufficient to confine oneself to dividing a territory by the soil type and restrict the research depth to 30 cm.

Keywords: *Semipalatinsk Test Site (STS), nuclear weapon tests, man-made radionuclides, migration of radionuclides, ecosystem contamination.*

INTRODUCTION

As we know, during nuclear tests, soil is the major ecosystem component exposed to contamination. It retains all substances that entered the ecosystem including man-made radionuclides for a long time due to contamination. Therefore, research into the content and distribution of radionuclides in the soil cover of the Semipalatinsk Test Site (STS) allows the obtainment of predictive values to characterize migration features of radionuclides in soils. The pattern of the vertical distribution of radionuclides at the test site has to be taken into account in developing rehabilitation techniques for radioactively contaminated areas.

Over years of research undertaken in the STS territory (between 1949 and 1989), 456 nuclear weapons tests were conducted. Of these, 116 tests were conducted in the atmosphere at the 'Experimental Field' site. 86 tests were conducted in the air, and 30 – above the ground (aboveground tests) [1, 2, 3]. Tests resulted in the contamination of soil cover with man-made radionuclides. Both test places within the Experimental Field' site and areas of passing radioactive plumes became contaminated. At the same time, fallout plumes went beyond the testing site, and, in certain cases, beyond STS [4, 5, 6]. In areas themselves, in which no nuclear tests were conducted and through which no fallout plumes were passing, the fallout background was formed, which differs from that of the global fallout in the northern hemisphere [7]. The next type of tests conducted at STS are underground nuclear tests. These tests were conducted in two ways. In boreholes at the 'Balapan' and 'Sary-Uzen' sites and in horizontal mine workings – adits at the 'Degelen' site. In the first case, the soil cover in the test place was not radioactively contaminated, except for individual abnormal situations. In the latter case, soil contamination with radionuclides currently continues due to the carry-away of radionuclides from adit cavities via ground waters [8].

The soil cover in the STS territory became contaminated not only due to nuclear weapon tests but also due to damage effects of radiological warfare agents tested (RWA). Such tests in the STS territory were conducted within the '4' and '4A' sites [9]. The pattern of radioactive contamination at these sites is 'spotty' with the spot size from hundreds of meters to several kilometers.

The global scientific literature has available many materials that characterize features of the vertical migration of radionuclides. In particular, the migration of radionuclides in soils contaminated due to radiological accidents at nucelar fuel cycle enterprises (the radiological accidents in the Southern Urals in 1957, and in Pripyat town in 1986 (the Chernobyl NPP) has benn sufficiently studied [10, 11]. Most of the data on the recent fallout were obtained within the short period of time following the accidents. Whereas after testing in individual STS areas, more than 70 years have now passed. Some authors provide research findings on the relationship between features of the vertical migration of radionuclides in soil and its physical and chemical properties or source species of radionuclides in soils [12–15], as well as depending on landscape conditions [16–18] and the existing external natural factors [19]. Certain researchers cite data on the distribution of radionuclides in soils of forest ecosystems located within the territory adjacent to STS [20] and in the soil of STS areas that are beyond test site areas [21].

The objective of this article was to address the pattern of the vertical distirbution of radionuclides in the undisturbed soil of various STS areas long after nuclear weapons and nuclear effects of radiological warfare agents were tested to shape the concept of the migration of radionuclides in vertical profiles of STS soils.

Thus, because of a variety of existing areas that differ in soil contamination, the STS territory is of interest to research into the vertical migration of radionuclides in the soil. The soil of steppe landscapes at the 'Experimental Field', '4A' sites and areas of the passing fallout in the form of plumes are of the greatest interest because, when undertaking research, most of contaminated areas were not destructed. Thus, it is possible to observe the natural vertical distribution of radionuclides in the soil. Of special interest are radioactively contaminated meadow soils of the 'Degelen' site because of many farm enterprises surrounding it, engaged in farm animal grazing and procurement of vegetative forage around this testing site.

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1 MATERIALS AND TECHNIQUES

1.1 Sampling

In terms of soil and geography, the test site territory is included in the steppe zone and is mostly represented by the chestnut soil type.

To research into the vertical distribution of man-made radionuclides, soil was sampled layerwsie to be analyzed for radionuclides. The sampling area of each layer was 200 cm^2. A trowel (preset geometry) was used for sampling (5×10×10 cm). Samples were collected layerwise from the following areas:

− in the soil cover of steppe ecosystems from places of aboveground and air nuclear tests at the 'Experimental Field' site (8 sampling points). The sampling interval – 3 cm as deep as 60 cm. Soil was sampled layerwsie next to ground zeroes of aboveground explosions with maxima of γ- and β – indicators, from areas with no visible man-made disturbance of the soil cover.

− in the fallout plumes that resulted from passing radioactive clouds of explosions conducted on August 12, 1953, May 20, 1960 and July 1, 1961 (8 sampling points).

− in RWA test places at the '4A' testing site (5 sampling points). Soil was sampled layerwise in places of maxima of β–indicators determined as a result of the previous radiometric survey.

− in meadow ecosystems of the 'Degelen' testing site associated with radioactively contaminated water streams from adits No. 176 and 177 (2 sampling points).

− in conventionally background STS areas (47 sampling points). The sampling interval – 3 cm as deep as 30 cm and further at 5 cm intervals from 30 to 50 cm deep. Stratified soil sampling points were selected on chestnut soils including the main types, subtypes and geni. This is attributed to the fact that the STS territory is represented by chestnut soils. Individual stratified sampling points were selected on saline soils.

1.2 Radionuclide analysis

Activity concentrations of man-made isotopes studied were determined as per guidelines [22, 23, 24] with a calibrated equipment that was entered in the State Register of Measuring Instruments of the Republic of Kazakhstan. Activity concentrations of ^{137}Cs and ^{241}Am were determined using a Canberra GX-2020 γ-spectrometer. The activity concentration of ^{90}Sr was determined following the radiochemical separation with a β-spectrometer, model TRI-CARB 2900 TR. The content of $^{239+240}$Pu was determined following the radiochemical separation with an α-spectrometer by Canberra. The detection limits were as follows: for ^{137}Cs – 4 Bq/kg, ^{241}Am – 1 Bq/kg, ^{90}Sr –5 Bq/kg, $^{239+240}$Pu –1 Bq/kg. The measurement uncertainty did not exceed the following precentage values: for ^{137}Cs and ^{241}Am – 20%, ^{90}Sr – 25%, $^{239+240}$Pu – 30 %.

1.3 Expression of results

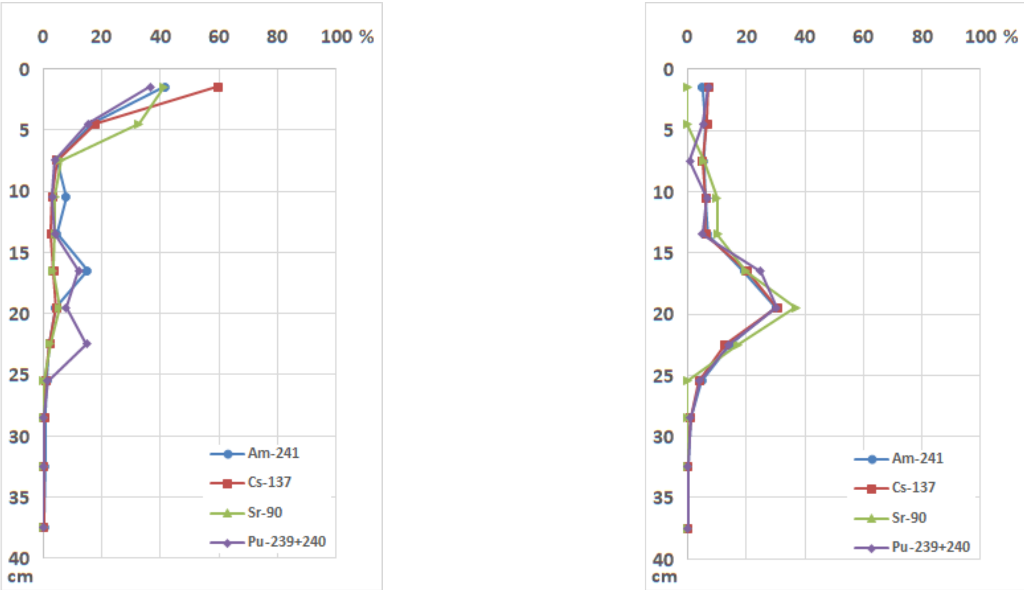
Activity concentrations of radionuclides in the soil are not provided. The paper presents processed figures as a percentage of the total activity of radionuclides in the soil profile. This presentation makes it possible to address the vertical distribution of radionuclides in the soil as aimed by this paper.

2 RESULTS AND DISCUSSION

2.1 Vertical distribution of radionuclides in the soil of the 'Experimental Field' testing site

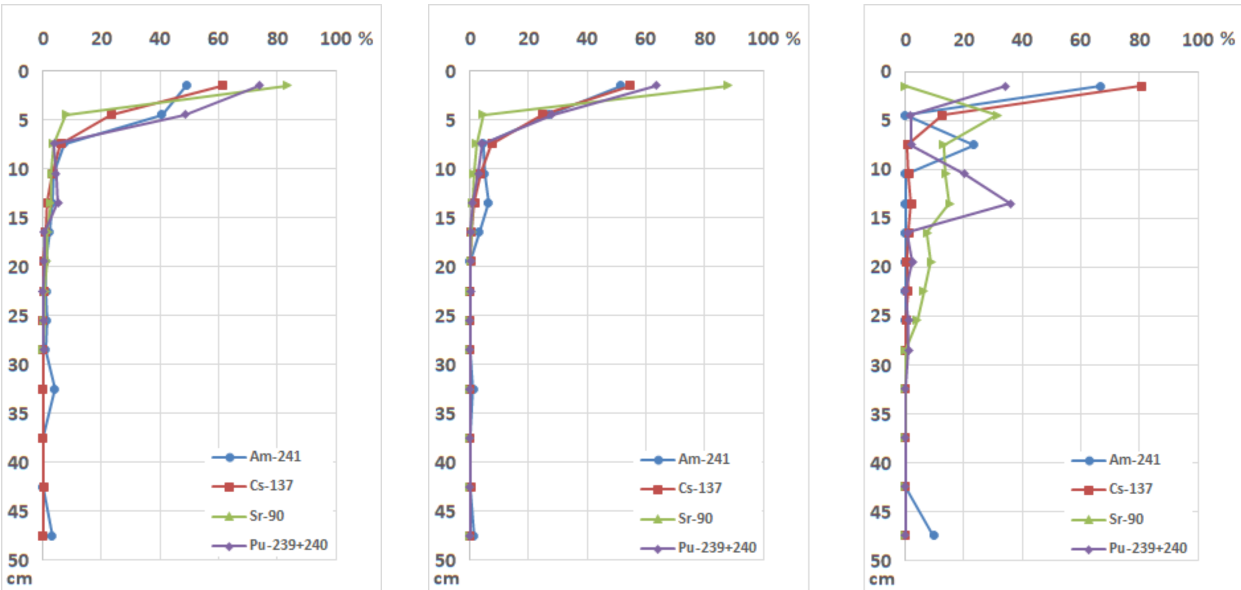
The figure (Figure 1) shows the vertical distribution of radionuclides typical of the 'Experimental Field' site, at which aboveground tests of nuclear weapons were conducted. One can see that the highest content of radionuclides is noted as deep as 5 cm followed by a sharp decrease. (Figure 1, a). The content of radionuclides in soil layers deeper than 15 cm is observed to jump. For instance, in one area, the maximum content of all radionuclides is at about 20 cm (Figure 1, b). The assumption was made that, at this point, the soil cover was disturbed due to anthropogenic activities. Analyses of physical and chemical soil properties showed that the humus content in lower layers is higher than in the first from the surface. The content of carbonates and soil density for all soil layers were also at the same level. These facts prove that the soil cover integrity has been disturbed.

Thus, in areas of aboveground nuclear tests at the 'Experimental Field' site, if there are no signs of disturbed soil cover, the vertical distribution of radionuclides in the soil is classical when maxima are noted for topsoil followed by a decrease. The low migration ability of radionuclides is explained by the fact that they are mainly in a tightly bound form [25].



a) b)

Figure 1. Distribution of radionuclides at the 'Experimental Field' testing site: a) average distribution based upon data from 8 areas, b) an area of disturbed soil cover



a) b) c)

Figure 2. Vertical distribution of radionuclides in the soil of fallout plumes: a) average distribution of radionuclides based upon data from 8 stratified sampling points; b) distribution of radionuclides characteristic of light-chestnut crushed stony soils in bald peak slopes; c) distribution of radionuclides in solonetzic-saline soils

2.2 Vertical distribution of radionuclides in the soil of fallout plumes

The vertical distribution of radionuclides in the soil cover of fallout plumes is depicted in the figure (Figure 2). Similarly to previous types of radioactive contamination, maximum activity concentrations of radionuclides are registered as deep as 10 cm. (Figure 2, a). At the same time, the maximum content of radioactive isotopes is noted as deep as 5 cm in bald peak slopes, represented by light-chestnut crushed stony soils (Figure 2, b). And in depressions between bald peaks, represented by friable solonetzic-saline soils, radionuclides are observed to transfer more to lower soil layers (Figure 2, c).

2.3 Vertical distribution of radionuclides in soils of the '4A' site

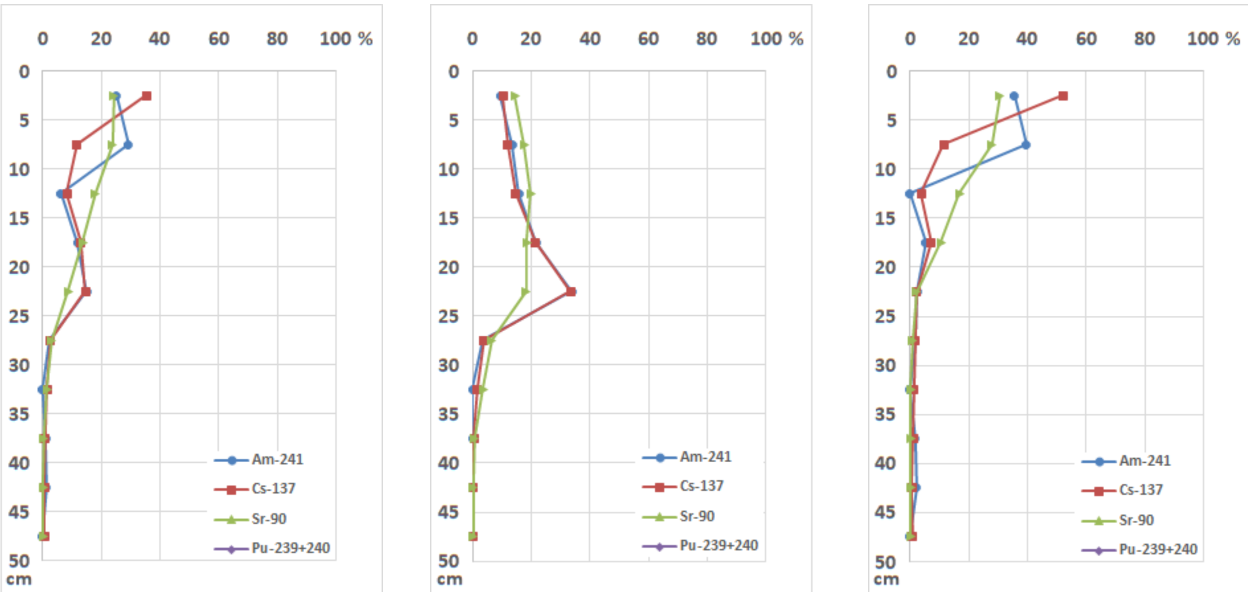
The vertical distribution of radionuclides in soils of the '4A' site is depicted in the figure (Figure 3). One can see that the distribution of radionuclides is different from the previous contamination types under discussion. ^{90}Sr is observed to be more mobile in the vertical soil profile, which is attributed to the mobility of this isotope as a whole and to soils with a light particle-size composition at the '4A' site [26]. The vertical distribution of ^{137}Cs and ^{241}Am in soils of the '4A' site differs greatly from their distribution in previous areas. For example, in area No. 2, maxima of activity concentrations of man-made radionuclides ^{137}Cs, ^{241}Am and ^{90}Sr are observed 22 cm deep (Figure 3, b), and in area No. 24 – in the 17 cm layer (Figure 3, c). In either case, a classical decrease in activity concentrations of radionuclides with depth is noted after activity concentration peaks. Thus, with this type of tests, one can assume two migration mechanisms of radionuclides. The first mechanism is traced from the topsoil to the layer with maxima of activity concentrations of radionuclides. At the same time, this soil layer acts as a geochemical barrier to the radioactive solution that is spilled at the time of testing and penetrating as far as the barrier layer by gravity and precipitation. The example is depicted in area 2 (Figure 3, b, c).

The second mechanism can be observed from the layer acting as a geochemical barrier, from which a classical distribution of radionuclides is noted (Figure 3, b).

2.4 Vertical distribution of radionuclides in soils of the 'Degelen' site

The figure (Figure 4) illustrates the distribution of radionuclides in the vertical soil profile of areas linked to water streams from adits. These areas are additionally moistened by water streams from test adits. A high mobility of radionuclides is attributed to the existing additional moistening. For example, maxima of the activity concentration of ^{241}Am are noted at a depth of 100 cm. A sufficiently high fate of ^{90}Sr and $^{239+240}$Pu is also noted at this depth. With no additional moistening, the vertical distribution of isotopes in the soil can be defined as classical.

The deep penetration of radionuclides is attributed not only to the presence of additional moistening in the form of creeks but also to the ongoing carry-away of radionuclides via adit waters [27]. Water monitoring carried out in adits of the 'Degelen' testing site in 2003 showed that the annual average concentration of ^{137}Cs in the water exceeded the intervention level (11 Bq/kg) by 7 to 47 times. Research undertaken by authors earlier demonstrated that in the water stream from adit 176, the activity concentration of ^{137}Cs reached 68 Bq/kg and that of ^{90}Sr – 155 Bq/kg. Maxima of the activity concentration of $^{239+240}$Pu were registered in the water stream from adit No. 503, where the concentration exceeded level of intervention equal to 0.56 Bq/kg by 12 times [28].



a) b) c)

Figure 3. Vertical distribution of radionuclides in soils of the '4A' site: a) average vertical distribution of radionuclides based upon data from 5 sampling points; b) vertical distribution of radionuclides in area 2; c) vertical distribution of radionuclides in area 24

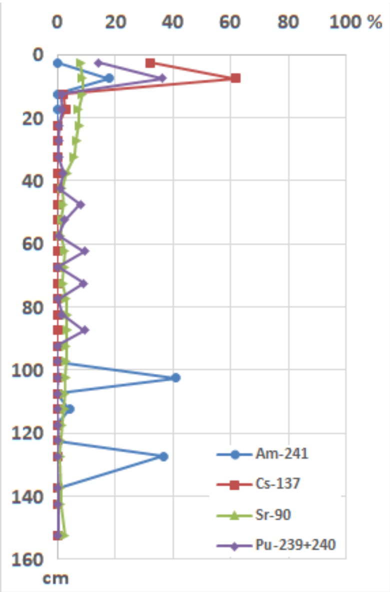


Figure 4. Vertical distribution of radionuclides in water-saturated meadow soils

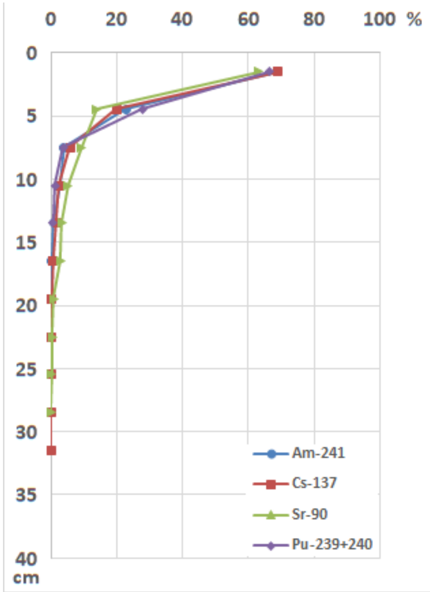


Figure 5. Vertical distribution of radionuclides in the soil of conventionally 'background' areas

Table. Functions describing vertical distribution curves of radionuclides in the soil

Subtype, type of soil	^{137}Cs	^{90}Sr	^{241}Am	$^{239+240}$Pu
Light-chestnut subtype	$y=(102\pm11)e^{-(0.3\pm0.03)}$	$y=(47\pm10)e^{-(0.2\pm0.05)x}$	$y=(76\pm16)e^{-(0.3\pm0.04)x}$	$y=(90\pm16)e^{-(0.4\pm0.05)x}$
Chestnut subtype	$y=(85\pm22)e^{-(0.4\pm0.08)x}$	$y=(51\pm16)e^{-(0.2\pm0.08)x}$	$y=(106\pm14)e^{-(0.4\pm0.04)x}$	$y=(128\pm28)e^{-(0.4\pm0.07)}$
Meadow-chestnut subtype	$y=(85\pm12)e^{-(0.3\pm0.03)x}$	$y=(49\pm11)e^{-(0.2\pm0.09)x}$	$y=108e^{-0.3x}$	$y=34e^{-0.2x}$
Chestnut subtype	$y=(125\pm13)e^{-(0.4\pm0.04)x}$	$y=(79\pm7)e^{-(0.3\pm0.04)x}$	$y=(85\pm12)e^{-(0.3\pm0.03)x}$	$y=(126\pm17)e^{-(0.4\pm0.04)x}$
Saline type	$y=(107\pm19)e^{-(0.4\pm0.05)x}$	$y=(24\pm5)e^{-(0.07\pm0.06)x}$	n/a	$y=(87\pm16)e^{-(0.4\pm0.008)x}$

2.5 Vertical distribution of radionuclides in the soil of the conventionally 'background' STS territory

The vertical distribution of radionuclides in the soil of the conventionally 'background' STS territory is depicted in the figure (Figure 5). Our paper [21] illustrates that in the conventionally 'background' territory, that is the one in which no nuclear tests were conducted, radioactive isotopes are concentrated in the topsoil as deep as 5 cm in various subtypes and variants of chestnut soils under arid conditions with no additional moistening.

Being formed under conditions of intense hydrothermal regime and high shortage of moisture that is only received by precipitation, zonal chestnut and light-chestnut soils largely differ in shallow wetting from the surface. Therefore, the vertical distribution of radionuclides in different soil variants and subtypes does not differ much. Functions that describe the pattern of the vertical distribution of radionuclides in subtypes of chestnut soils are very similar to one another (see Table) [13].

Exceptions can be meadow-chestnut soils. Differences between functions derived for the chestnut soil type and the ones derived for saline soils can be noted. These are the same for radioactive isotopes ^{137}Cs and $^{239+240}$Pu, whereas for ^{90}Sr these functions are greatly different.

Thus, taking into account the poor differentiation of the distribution pattern of radionuclides, possibly related to the measurement uncertainty, methodological uncertainties in the field work and factors ignored that cannot be identified at the time of sampling (mixing of layers by burrowers or farm animals etc.), one can state that it is sufficient to classify the soil cover by the soil type for assessing the distribution of radionuclides in the vertical soil profile of the STS territory.

Besides, radionuclides in soils of 'background' areas not referring to testing sites, are concentrated in the topsoil and are mostly detectable as deep as 15–20 cm. Thus, this fact allows the optimization of the survey of STS areas by restricting the research depth to 30 cm (provided there is no additional moistening by water streams or signs of disturbed soil cover).

Conclusion

Under arid conditions, without additional moistening of soils, the maximum concentration of artificial radionuclides is only in the 0–5, 5–10 cm topsoil.

In places of radiological warfare agent tests at '4A' site, two zones with different mechanisms of the vertical distribution of ^{137}Cs, ^{241}Am, ^{90}Sr and $^{239+240}$Pu artificial radionuclides were identified. The first zone spreads to the depth at which radionuclides were initially distributed with the one-time spill of a radioactive liquid at local points, immediately at the moment of tests and further penetration of the recent fallout with precipitation deep down the soil to some definite soil (buffer) layer. The second zone is the zone of classically distributed radionuclides from a soil (buffer) layer to its lower layers.

Artificial disturbances of the soil cover that took place during the nuclear tests and preparatory activities, can disturb the common pattern of radionuclides distribution in the soil profile.

In case soils receive additional moistening, due to either surface flow or ground water, an intensive migration of artificial radionuclides, first of all, that of ^{90}Sr, as the best soluble and the most mobile radionuclide, is noticed.

Research findings show that to assess the distribution of radionuclides in vertical profiles of STS soils, it is sufficient to classify the soil cover by the soil type.

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СЕМЕЙ СЫНАҚ ПОЛИГОНЫ ТОПЫРАҚТАРЫНДА РАДИОНУКЛИДТЕРДІҢ ТІГІНЕН ТАРАЛУЫ

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Мақалада Семей сынақ полигоны топырағындағы негізгі ұзақ өмір сүретін ^{137}Cs, ^{241}Am, ^{90}Sr және $^{239+240}$Pu техногендік радионуклидтерінің вертикалды таралу сипаты қарастырылған. Радионуклидтік ластанудың әр түрлі сипаты бар учаскелер қаралды – «Тәжірибе даласы» сынақ алаңында ядролық және термоядролық қаруға жер үсті сынақтарын жүргізу орындары, Семей сынақ полигоны аумағының шегінде іздер түріндегі радиоактивті түсулер, «4а» сынақ алаңында орналасқан әскери радиоактивті заттектерді сынау учаскелері, «Дегелең» сынақ алаңының сынақ штольняларынан радиоактивті-ластанған су ағындарымен жанасқан шалғынды экожүйелер учаскелері, тікелей ядролық және термоядролық каруларға сынақтар жүргізілмеген полигонның шартты «фондық» аумақтары. Зерттеу барысында зерттелетін радионуклидтердің көрсетілген учаскелердің топырақтарындағы тереңдігі бойынша таралу сипатындағы айырмашылықтар анықталды. Айырмашылықтар радионуклидтік ластанудың пайда болу сипатына және топырақтың физика-химиялық қасиеттері, ылғалдандыру режимі, адам іс-әрекеті және т.б. сияқты абиотикалық және биотикалық факторлардың болуына байланысты. Алынған деректер негізінде бұрынғы Семей сынақ полигонының топырақ жамылғысында радионуклидтердің вертикалды таралуын зерттеуді оңтайландыруға бағытталған ұсыныстар әзірленді. Атап айтқанда, мұндай зерттеулер жүргізу кезінде топырақты топырақ түріне жіктеп, зерттеуді 30 см тереңдікке дейін шектеу жеткілікті екендігі анықталды.

Түйін сөздер: *Семей сынақ полигоны (ССП), ядролық қаруды сынау, техногендік радионуклидтер, радионуклидтердің жылыстауы, экожүйелердің ластануы.*

ВЕРТИКАЛЬНОЕ РАСПРЕДЕЛЕНИЕ РАДИОНУКЛИДОВ В ПОЧВАХ СЕМИПАЛАТИНСКОГО ИСПЫТАТЕЛЬНОГО ПОЛИГОНА

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В статье рассмотрен характер вертикального распределения основных долгоживущих техногенных радионуклидов ^{137}Cs, ^{241}Am, ^{90}Sr и $^{239+240}$Pu в почвах Семипалатинского испытательного полигона. Рассмотрены участки с различным характером радионуклидного загрязнения – места проведения наземных испытаний ядерного и термоядерного оружия на испытательной площадке «Опытное поле», радиоактивные выпадения в виде следов в пределах территории Семипалатинского испытательного полигона, участки испытания боевых радиоактивных веществ, расположенные на испытательной площадке «4А», участки луговых экосистем, сопряженных с радиоактивно-загрязненными водотоками из испытательных штолен испытательной площадки «Дегелен», условно «фоновые» территории полигона, непосредственно на которых испытаний ядерного и термоядерного оружия не проводилось. В ходе исследований выявлены различия в характере распределения исследуемых радионуклидов по глубине в почвах указанных участков. Различия обусловлены характером образования радионуклидного загрязнения и наличием абиотических и биотических факторов, таких, как, физико-химические свойства почв, режим увлажнения, деятельность человека, и др.. На основании полученных данных разработаны рекомендации, направленные на оптимизацию исследований вертикального распределения радионуклидов в почвенном покрове бывшего Семипалатинского испытательного полигона. В частности, установлено, что при проведении таких исследований достаточно классифицировать почву до типа почвы и ограничить исследования до глубины 30 см.

Ключевые слова: *Семипалатинский испытательный полигон (СИП), испытания ядерного оружия, техногенные радионуклиды, миграция радионуклидов, загрязнение экосистемы.*