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General Methodology for Assessing the Content of Artificial Radionuclides in Livestock Products Produced in Areas Polluted by Nuclear Tests

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General Methodology for Assessment the Content of Artificial Radionuclides in Livestock Products Produced in Areas Polluted by Nuclear Tests

Abstract

The Semipalatinsk Test Site (STS) in Kazakhstan, active from 1949 to 1989, resulted in significant radioactive contamination. Following the site's closure, unauthorized agricultural activities led to potential human exposure to radionuclides through livestock products. This dissertation aims to assess the content of artificial radionuclides in livestock products produced in these contaminated areas, focusing on the transfer parameters of ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am from the contaminated environment into livestock and poultry.

The primary objective of this research is to investigate the distribution and transfer parameters of key radionuclides in agricultural animals and poultry under the conditions of the STS. The study involves examining the dynamics of radionuclide transition into animal organs and tissues, determining transfer parameters from feed, soil, and water into various livestock species, and studying the transfer parameters into poultry products.

The research was conducted at the STS territory. Experimental subjects included pregnant mares, yearling fillies, and 50-day-old broilers. These animals were exposed to contaminated feed, soil, and water to study the transfer of radionuclides into their tissues.

The findings revealed significant radionuclide transfer into various animal tissues. Studies on horses indicated that younger animals, such as yearling fillies, exhibited higher transfer parameters for ^{137}Cs and ^{90}Sr compared to adults. Additionally, the concentration of $^{239+240}$Pu and ^{241}Am in foetal tissues was measured, providing insights into potential risks for offspring.

Research on broilers demonstrated that the dynamics of radionuclide accumulation varied over time, with ^{137}Cs and ^{241}Am showing different patterns of

distribution in muscle, liver, and bone tissues. Transfer coefficients (F_f) and concentration ratios (C_R) for broilers' meat were established, offering critical data for assessing human dietary exposure.

The findings underscore the need for ongoing monitoring and assessment of radionuclide contamination in livestock products in the STS area. The transfer parameters derived from this study are essential for developing predictive models to evaluate the risks associated with consuming contaminated livestock products. Additionally, the research provides a scientific basis for formulating guidelines and policies to manage and mitigate the impacts of radioactive contamination on agriculture and public health.

Key findings of the research include: $^{239+240}$Pu and ^{241}Am showed specific distribution patterns in animal tissues, with the liver being the primary site of accumulation. Transfer coefficient values varied significantly based on the contamination source and type of feed, highlighting the complexity of radionuclide transfer in different environmental contexts. Younger animals exhibited higher transfer parameters for ^{137}Cs and ^{90}Sr, indicating age-dependent differences in radionuclide accumulation. The study provided crucial data on the transfer of ^{137}Cs and ^{241}Am into broiler tissues, essential for assessing the safety of poultry products from contaminated areas.

In conclusion, this research provides a critical assessment of radionuclide transfer in livestock and poultry, offering vital insights for managing the impacts of nuclear contamination on agriculture and public health. The findings support the development of robust risk assessment models and inform strategies for mitigating radiation exposure in contaminated regions.

Ядролық сынақтармен ластанған аймақтарда өндірілетін мал шаруашылығы өнімдеріндегі жасанды радионуклидтердің құрамын анықтаудың жалпы әдістемесі

Түйіндеме

1949 жылдан 1989 жылға дейін жұмыс істеп тұрған Қазақстандағы Семей сынақ полигоны (ССП) айтарлықтай радиоактивті ластануға алып келді. Полигон жабылғаннан кейін рұқсат етілмеген ауылшаруашылық қызметі мал шаруашылығы өнімдері арқылы радионуклидтердің адамға ықтимал әсеріне әкелді. Бұл диссертацияның мақсаты перехода ^{137}Cs, ^{90}Sr, $^{239+240}Pu$ және ^{241}Am өту параметрлеріне ерекше назар аудара отырып, осы ластанған аумақтарда өндірілетін мал шаруашылығы өнімдеріндегі жасанды радионуклидтердің құрамын бағалау болып табылады.

Бұл зерттеудің негізгі мақсаты ССП жағдайында ауылшаруашылық жануарлары мен құстардың негізгі дозалаушы радионуклидтердің таралу және ауысу параметрлерін зерттеу болып табылады. Зерттеуге радионуклидтердің Жануарлар мүшелері мен тіндеріне ауысу динамикасын зерттеу, жемнен, топырақтан және судан малдың әртүрлі түрлеріне ауысу параметрлерін анықтау және құс өнімдеріне ауысу параметрлерін зерттеу кіреді.

Зерттеу ССП аумағында жүргізілді. Эксперимент нысандары ретінде жүкті биелер, бір жасар биелер және 50 күндік бройлер тауықтары пайдаланылды. Бұл жануарлар тіндеріндегі радионуклидтердің тасымалдануын зерттеу үшін ластанған Жемге, топыраққа және суға ұшырады.Нәтижелер радионуклидтердің жануарлардың әртүрлі ұлпаларына айтарлықтай ауысуын көрсетеді.

Жылқыларға жүргізілген зерттеулер көрсеткендей, бір жасар филли сияқты жас жануарлар ересектермен салыстырғанда ^{137}Cs және ^{90}Sr тасымалдау көрсеткіштерін жоғарылатады. Сонымен қатар, ұрық тіндеріндегі $^{239+240}Pu$ және ^{241}Am концентрациясы өлшенді, бұл ұрпақ үшін ықтимал қауіптер туралы түсінік берді.

Бройлер тауықтарындағы зерттеулер радионуклидтердің жинақталу динамикасы уақыт өте келе өзгеретінін көрсетті, бұлшықет, бауыр және сүйек тіндерінде ^{137}Cs және ^{241}Am әртүрлі бөлінеді. Бройлер тауықтарының еті үшін тасымалдау коэффициенттері (F_f) және концентрациялары (C_R) анықталды, бұл адам ағзасына диеталық әсерді бағалау үшін маңызды мәліметтер берді.

Алынған нәтижелер ССП аймағында мал шаруашылығы өнімдерінің радионуклидтермен ластануын тұрақты мониторингтеу және бағалау қажеттігін көрсетеді. Осы зерттеу нәтижесінде алынған тасымалдау параметрлері ластанған мал өнімдерін тұтынумен байланысты тәуекелдерді бағалау үшін болжамды үлгілерді әзірлеу үшін қажет. Сонымен қатар, зерттеу ауыл шаруашылығы мен қоғамдық денсаулық сақтау үшін радиоактивті ластануды басқару және азайту бойынша нұсқаулар мен саясатты әзірлеуге ғылыми негіз береді.

Зерттеудің негізгі нәтижелері мыналарды қамтиды: $^{239+240}Pu$ және ^{241}Am Жануарлар тіндерінде таралудың нақты заңдылықтарын көрсетті, негізгі жинақтау орны бауыр болып табылады. Тасымалдау коэффициентінің мәндері ластану көзіне және жем түріне байланысты айтарлықтай өзгерді, бұл әртүрлі экологиялық жағдайларда радионуклидтерді тасымалдаудың күрделілігін көрсетеді. Жас жануарларда ^{137}Cs және ^{90}Sr тасымалдау көрсеткіштері жоғары болды, бұл радионуклидтердің жинақталуындағы жас айырмашылықтарын көрсетеді. Зерттеу ластанған аймақтардан құс өнімдерінің қауіпсіздігін бағалау үшін қажет бройлер тіндеріндегі ^{137}Cs және ^{241}Am тасымалдау туралы маңызды деректерді алды.

Қорытындылай келе, бұл зерттеу радионуклидтердің мал мен құс арқылы тасымалдануына сыни баға береді, бұл ауыл шаруашылығы мен қоғамдық денсаулық сақтау үшін ядролық ластанудың әсерін басқару үшін маңызды ақпарат береді. Нәтижелер ластанған аймақтарда тәуекелдерді бағалаудың сенімді модельдерін және радиациялық әсерді азайту Стратегияларын жасауға көмектеседі.

Общая методология оценки содержания искусственных радионуклидов в продукции животноводства, производимой в районах, загрязненных ядерными испытаниями

Реферат

Семипалатинский испытательный полигон (СИП) в Казахстане, действующий с 1949 по 1989 год, привел к значительному радиоактивному загрязнению. После закрытия полигона несанкционированная сельскохозяйственная деятельность привела к потенциальному воздействию радионуклидов на человека через продукты животноводства. Целью данной диссертации является оценка содержания искусственных радионуклидов в продуктах животноводства, производимых на этих загрязненных территориях, с уделением особого внимания параметрам перехода ^{137}Cs, ^{90}Sr, $^{239+240}$Pu и ^{241}Am.

Основной целью данного исследования является изучение параметров распределения и перехода основных дозобразующих радионуклидов сельскохозяйственными животными и птицей в условиях СИП. Исследование включает изучение динамики перехода радионуклидов в органы и ткани животных, определение параметров переноса из кормов и почвы в органы и ткани лошадей, а также изучение параметров переноса в продукты птицеводства.

Исследование проводилось на территории СИП. В качестве объектов эксперимента использовались беременные кобылы, годовалые кобылки и 50-дневные цыплята-бройлеры. Полученные данные свидетельствуют о значительном переносе радионуклидов в различные ткани животных

Исследования на лошадях показали, что молодые животные, такие как годовалые кобылки, демонстрируют более высокие показатели переноса ^{137}Cs и ^{90}Sr по сравнению со взрослыми. Кроме того, была измерена концентрация $^{239+240}$Pu и ^{241}Am в тканях плода, что позволило получить представление о потенциальных рисках для потомства.

Исследования на цыплятах-бройлерах показали, что динамика накопления радионуклидов меняется с течением времени, при этом ^{137}Cs и ^{241}Am по-разному распределяются в мышечной, печеночной и костной

тканях. Были установлены коэффициенты переноса (F_f) и концентрации (C_R) для мяса цыплят-бройлеров, что позволило получить важные данные для оценки воздействия на организм человека с пищей.

Полученные результаты подчеркивают необходимость постоянного мониторинга и оценки загрязнения радионуклидами животноводческой продукции в зоне СИП. Параметры переноса, полученные в результате этого исследования, необходимы для разработки прогнозных моделей для оценки рисков, связанных с потреблением загрязненной животноводческой продукции. Кроме того, исследование обеспечивает научную основу для разработки руководящих принципов и политики по управлению и смягчению последствий радиоактивного загрязнения для сельского хозяйства и общественного здравоохранения.

Основные результаты исследования включают в себя: $^{239+240}$Pu и ^{241}Am показали специфические закономерности распределения в тканях животных, причем основным местом накопления является печень. Значения коэффициента переноса значительно различались в зависимости от источника загрязнения и типа корма, что подчеркивает сложность переноса радионуклидов в различных условиях окружающей среды. У молодых животных показатели переноса ^{137}Cs и ^{90}Sr были выше, что указывает на возрастные различия в накоплении радионуклидов. В ходе исследования были получены важные данные о переносе ^{137}Cs и ^{241}Am в тканях бройлеров необходимые для оценки безопасности продуктов птицеводства из загрязненных районов.

В заключение, в этом исследовании дается критическая оценка переноса радионуклидов домашним скотом и птицей, что дает важную информацию для управления последствиями ядерного загрязнения для сельского хозяйства и общественного здравоохранения. Полученные результаты помогают разработать надежные модели оценки рисков и стратегии снижения радиационного воздействия в загрязненных регионах.

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List of Abbreviations and acronyms

STS – Semipalatinsk Test Site

IAEA – International Atomic Energy Agency

F_f – Transfer Coefficients

C_R – Concentration Ratio

AC – Activity Concentration

MPL – Maximum Permissible Levels

PL – Permissible Level

Bq kg^{-1} – Becquerels per Kilogram

Bq l^{-1} – Becquerels per liter

MBq – Megabecquerel

kBq – Kilobecquerel

mg kg^{-1} – Milligrams per Kilogram

$^{239+240}$Pu – Plutonium–239+240

^{241}Am – Americium–241

^{137}Cs – Cesium–137

^{90}Sr – Strontium–90

tab. – Table

fig. – Figure

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CHAPTER 1. INTRODUCTION

The Semipalatinsk Test Site (STS) was active in East part Kazakhstan from 1949 to 1989. Today, the STS has localised areas with high concentrations of radionuclides (Lukashenko et al., 2015). After the test site was closed, the local population began unauthorized commercial activities in the territory of the STS. More than one hundred farms are currently operating year-round in the STS. These farms typically graze, and breed sheep, cattle and horses (including meat for human consumption). Horse meat is a relatively important animal derived food product in Kazakhstan (Nyssanbayev, 1998).

The total area of the STS is about 18 thousand square kilometers, with over 90% of the territory exhibiting “background” radiation levels (Lukashenko et al., 2010). The formal transition of these areas for commercial utilization is currently in progress. To facilitate this process and ensure public health, it is crucial to make scientifically based predictions of radionuclide activity concentrations in animal-derived foodstuffs. Robust dose predictions for consumers are essential tools in achieving this aim, providing a foundation for safe commercial use of the land.

Knowledge on the transfer of ^{137}Cs and ^{90}Sr radionuclides to livestock products is relatively available (Beresford & Howard, 2011; Howard et al.2009, Fesenko et al. 2007, 2009) from both laboratory and field studies. Feed to animal product transfer parameters have been compiled in international recommendations (IAEA 2010). However, the STS is considerably different compared to the previous studies in terms of local conditions and source terms. The review of Beresford et al. (2000) demonstrates that source-dependent radiocesium bioavailability is one of the major parameters determining the degree of contamination in the milk and meat of ruminant livestock. Furthermore, there are relatively few studies of the transfer of *Pu* and *Am* to livestock (IAEA, 2010) and these radionuclides are important contaminates within the STS.

In the STS the major source of ingested radionuclides for livestock is contaminated soil; the unintentional daily consumption is estimated to be

approximately 1.6 kg per day for horse and cattle and 0.2 kg per day for sheep in the pasturage period (Lukashenko et al., 2015). It has been reported that the amount of soil consumption by cattle can reach 18% of the dry matter daily intake of forage (Wilkins et al., 1997). For *Cs* ingested soil has been demonstrated to have a lower bioavailability than contaminated forage, the Transfer Coefficient (ratio of Radionuclide activity concentration in animal tissue (Bq kg^{-1}fresh mass) / daily intake of the radionuclide, Bq day^{-1}) for ^{137}Cs could differ from threefold up to one order of magnitude, for *Pu* from fivefold up to 2 orders of magnitude (Beresford et al. 2000). The local conditions significantly influence Transfer Coefficients, and the source of contamination, one singular value can't be applied for the whole STS area.

Comparatively few studies have considered the transfer of radionuclides to horses. In a study conducted in the STS, Semioshkina et al. (2006) reported the transfer of ^{137}Cs and ^{90}Sr into the milk and tissues of horses. Isamov et al. (2009) and Tsygvintsev et al. (2004) reported studies of ^{137}Cs accumulation and excretion in horsemeat after the Chernobyl accident.

There are few studies considering the transfer of actinides to animal derived food products using realistic dietary sources (Averin. et al. 2011; Howard et al. 2007; Beresford et al. 2007a). The review by Fesenko et al. (2018) reports on the transfer of radionuclides from various ingested materials to animal meat in the Russian language studies. There is some rare data presented for transuranic isotopes, however the bulk of the available material concerns only radioactive *Cs* and *Sr*. Estimated *Pu* transfer coefficients for milk and liver from these dietary sources are in the range 10^{-6} to 10^{-5} d kg^{-1} and 10^{-3} to 10^{-2} d kg^{-1}, respectively. Whilst studies have estimated transfer parameter values for $^{239+240}Pu$ and ^{241}Am to mutton for the STS (Baigazinov et al., 2013, Panitskiy et al., 2011), the lack of data for locally important horse products impacts on our ability to conduct robust risk assessments.

After analyzing the main publications on current world experiences (IAEA, 2009, 2010; Green, 2003; Fesenko, 2018 & 2019, Howard, 2009) we found that

there is no available data for *Am* in poultry meat. Few data sets are available for estimating the transfer of radionuclides in "soil-products" systems (*Am*aral, 1995; Ng et al., 1982) as the soil can lead to radioactive contamination of farm products (Thornton & Abrahams, 1983; Beresford & Howard, 1991). The review of the Russian language studies by Fesenko et al. (2018) reports that the transfer of ^{137}Cs, ^{90}Sr, ^{54}Mn, ^{65}Zn to chicken tissue was described in four publications. The Transfer Coefficient F_f values for ^{137}Cs varies from 0.7±0.1 (Mean±SD) to 2.3±0.3 in different tissues for a duration of 30-360 days. The generated concentration ratios (C_R) of *Am* for the meat of different animals has been estimated as 1×10^{-4}. Moreover, it is a fact that the migration of the radionuclide into meat or milk will significantly vary depending on the source of intake. For instance, forage contaminated by *Cs* has a higher bioavailability than ingested soil (Beresford et al. 2000).

The STS presents a unique and significant challenge due to its extensive radioactive contamination from past nuclear tests. Understanding the transfer of radionuclides such as ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am to livestock products is crucial for ensuring the safety of food products and public health. This research highlights the importance of considering local environmental conditions and the sources of contamination, as these factors significantly influence the transfer coefficients and the bioavailability of radionuclides. The study underscores the need for robust, site-specific data to make accurate risk assessments and develop effective guidelines for the safe utilization of the STS territory for agricultural purposes. By addressing these concerns, we can better manage the risks associated with consuming contaminated livestock products and protect the health of populations in affected areas.

Research Objective:

The primary objective of this PhD thesis is assessing the content of artificial radionuclides in livestock products, specifically focusing on areas polluted by nuclear tests. This aims to enhance the understanding of radionuclide transfer to agricultural products, thereby improving radiological safety assessments and risk

management strategies for populations consuming livestock from contaminated regions by atomic bombs and could be extended for regions influenced by nuclear accidents.

Research Tasks:

1. Determine the transfer parameters of the radionuclides $^{239+240}$Pu, ^{241}Am, ^{137}Cs, and ^{90}Sr to the tissues of horses, specifically to quantify their transfer into the organs of horses with different ages and through the placental barrier into foetuses after feeding with contaminated soil or diet contaminated by a leachate solution.

2. Investigate the transfer parameters of radionuclides ^{241}Am and ^{137}Cs to the tissues of broilers' organs, with the aim of determining the Transfer Coefficient (F_f) and concentration ratios (C_R) for long-term intake of radionuclides from grass meal and soil.

3. Investigate the dynamics of accumulation and excretion of radionuclides ^{241}Am and ^{137}Cs in the muscle, liver, and bone of broilers after a long-term feeding experiment with contaminated soil and grass meal.

4. Evaluate the feasibility of livestock farming at the STS by analyzing regulatory standards, permissible radionuclide levels, agricultural practices, and radionuclide intake by livestock.

CHAPTER 2 HISTORICAL AND ENVIRONMENTAL OVERVIEW OF THE STS

2.1 The STS and Its Key Test Areas

The STS, located in northeastern Kazakhstan, covers approximately 18,300 square kilometers. It was used by the Soviet Union for nuclear testing from 1949 to 1989, conducting 456 nuclear tests. These tests included atmospheric, surface, and underground detonations. Key testing areas within STS included the Experimental Field, Degelen Mountain, Balapan, Sary-Uzen, Telkem and "4a". The Experimental Field hosted 116 atmospheric tests. Degelen Mountain saw 209 underground tests, while Balapan hosted 105 underground tests, including the creation of the artificial "Atomic Lake." Sary-Uzen and Telkem conducted 24 and 2 tests, respectively. The "4a" site was among the locations for testing solid or liquid radioactive substances, with 175 blasts employing conventional chemical explosives performed here. (Lukashenko, 2020). Currently, the STS remains heavily contaminated with radionuclides such as ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am.

Experimental Field was a key location for nuclear tests conducted by the USSR from 1949 to 1962. This site hosted both atmospheric and surface nuclear tests, with a total of 116 tests aimed at studying the effects of nuclear detonations. The area, covering approximately 375 km², is characterized by a flat landscape surrounded by low mountains. The radioactive contamination in this field varies, with high concentrations of radionuclides like ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am found at the epicenters of detonations and along fallout plumes. The contamination sites are localized, with specific areas showing extremely high activity. The activity concentration of transuranic radionuclides, such as $^{239+240}$Pu, can reach up to 100 kBq kg^{-1} of soil in areas less than 1 km². Similarly, the activity concentration of ^{137}Cs over 2 kBq kg^{-1} and ^{241}Am over 10 kBq kg^{-1} of soil in areas less than 1.5 km². (Moshkov et al. 2011, Kunduzbayeva et al. 2022)

"4a" Site was used for testing solid or liquid radioactive substances, employing conventional chemical explosives in 175 blasts. This area, covering

approximately 63 km², is part of the larger STS complex and is characterized by scattered contamination zones where various radioactive materials were tested. The "4a" site shows elevated levels of radionuclides such as ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu, reflecting the diverse range of substances and experimental conditions employed in the tests. The radionuclide ^{90}Sr, contributing primarily to the site's radioactive contamination, is mostly found in the soil in its exchangeable form, with a notably high level of water-soluble forms. ^{137}Cs is predominantly in a firmly bound form (up to 94%). The prevailing form of ^{241}Am is the mobile form. The primary content of $^{239+240}$Pu, as on other sites, is noted in the form of firmly bound forms. The contaminated soil areas with ^{90}Sr range from 50 to 100 kBq kg^{-1} over 0.09 km², from 100 to 500 kBq kg^{-1} over 0.09 km², and from 500 to 1000 kBq kg^{-1}over 0.06 km² (IRSE, 2012).

Degelen Mountain was a crucial site for underground nuclear tests conducted by the USSR from 1961 to 1989. Located in the southern part of the STS, this area, covering about 238 km², hosted 209 underground tests in tunnels designed for low-yield explosions. The site is characterized by its rugged terrain and numerous tunnels. Radioactive contamination in this area is significant, mainly with radionuclides such as ^{137}Cs and ^{90}Sr detected in groundwater and sediments, primarily near tunnel entrances where water seepage has mobilized these contaminants over time. ^{90}Sr is most mobile in the soils of the "Degelen" site, with over half of this radionuclide's content in an exchangeable form. Less mobile in the meadow soils are radionuclides ^{241}Am and ^{137}Cs.

Balapan was another key site within the STS, hosting 105 underground nuclear tests from the early 1960s onwards. This area, covering around 190 km², is notable for the creation of "Atomic Lake," an artificial reservoir formed by a 130-kiloton nuclear explosion in 1965. The radioactive fallout in this region is concentrated around the lake and its surrounding areas, reflecting the high-yield nature of the explosions conducted here, with significant contamination from radionuclides including ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am. The Atomic Lake, formed by a nuclear explosion at the STS, and the Shagan River, which flows from it, are

significant sources of radioactive contamination, particularly by ^{3}H. Tritium activity concentrations in the Shagan River can reach up to 350 kBq L^{-1}, especially near the Atomic Lake and in areas up to 10 km downstream (Aidarkhanov et al, 2011).

Sary-Uzen was utilized for 24 underground nuclear tests, focusing on strategic military applications such as testing nuclear warheads and devices. The region, covering approximately 120 km², is characterized by its arid, steppe-like landscape. The radioactive contamination at Sary-Uzen includes notable concentrations of ^{137}Cs and ^{90}Sr, especially in areas close to the detonation sites where fallout plumes have deposited significant amounts of radioactive material.

Telkem saw a total of 2 excavation tests aimed at exploring the use of nuclear explosions for large-scale earth-moving projects. These tests were part of the USSR's program to develop industrial applications for nuclear technology. The contamination in Telkem, covering about 20 km², includes high levels of ^{137}Cs, ^{90}Sr, and transuranic elements, primarily concentrated at the sites of the excavations.

Background Areas are regions not directly used for nuclear tests but still affected by radioactive contamination. These areas reflecting the spread of radioactive particles from global fallout and contamination from primary test sites. The Northern part of the background area is mainly contaminated by global fallout, while the Southeastern part is partially affected by fallout from the thermonuclear tests conducted at the Experimental Field. These areas exhibit varying levels of radionuclides such as ^{137}Cs, ^{90}Sr, and transuranic elements like $^{239+240}Pu$ and ^{241}Am, with concentrations not higher than kilobecquerels per kg. The radioactive plumes in these regions can stretch over 100 km in length and 10 to 14 km in width, showing significant contamination despite not being the primary test locations (Lukashenko S.N. 2010, 2011, 2012, 2013, 2014). These findings indicate severe radioactive contamination, necessitating ongoing monitoring and remediation efforts to address the environmental and health impacts (Gorlachev et al., 2019)

2.2 Climate and Topography

The STS is located within the East Kazakhstan, Pavlodar, and Karaganda regions, covering an area of 18,500 square kilometers. The region experiences a sharply continental climate, characterized by cold winters and hot, dry summers. Winters typically span from November to March, with temperatures ranging from -16°C to -20°C, occasionally dropping to -46°C or lower. Snow cover, which reaches heights of 20-50 cm, forms in mid-November and melts by early April. Summers are marked by temperatures between +24°C and +27°C, reaching up to +42°C. The annual precipitation ranges from 160 to 400 mm, placing the area in a zone of insufficient and unstable moisture. The STS features a varied topography with small hills, low mountains, and river valleys, with elevations ranging from 250 to 1085 meters. Wind patterns vary with southeastern winds dominating in winter and northern winds in summer. The area frequently experiences blizzards in winter and dust storms in summer.

2.3 Soil Cover

The soil cover at the STS is primarily composed of chestnut and light chestnut soils, which are typical of the semi-arid steppe region. These soils are distributed across two main subzones: the dry steppe with chestnut soils and the desert steppe with light chestnut soils. The boundary between these subzones generally follows the 50°20' - 50°30' latitude. In the central, southern, and eastern parts of the site, light chestnut soils predominate, while chestnut soils are more common in the northern, western, and southern areas. Soil heterogeneity is influenced by moisture conditions, relief, and the composition of parent materials. The soils include carbonate, solonetzic, and less-developed types. Meadow-chestnut soils form under meadow-steppe vegetation in low-lying areas, and salt-affected soils such as solonetz and solonchak are also present. Human activities, particularly nuclear testing, have significantly altered the soil properties, leading to variations in organic matter content and pH levels. These factors affect the migration and distribution of artificial radionuclides within the test site's environment.

2.4 Vegetation Contamination Characteristics at the STS

The vegetation cover across the majority of the STS is represented by desert-steppe vegetation on light-chestnut soils, characterized by a widespread presence of both steppe elements, such as turf grasses, and desert elements, including semi-shrub sages and, to some extent, saltworts.

The species composition of semi-desert communities is significantly poorer than that of steppes, with the vegetation cover of the semi-desert also featuring a high prevalence of sagebrush and a minimal amount of diverse steppe grasses. The average biological productivity of pastures is 2-4 centners per hectare. The area is characterized by dry steppe diverse grasses.

The flora diversity, the quantitative and qualitative assortment of species, genera, and families in the Degelen mountain range are typical for the flora of the eastern part of the Central Kazakh small hillock steppe. The cenotic composition of the vegetation cover of the Degelen mountain range is notably diverse. A distinctive feature of the vegetation structure of the mountain range is the altitudinal zonal vegetation change. This area is primarily represented by meadow diverse grasses, whose productivity can vary from 2 to 10 centners per hectare, depending on the location and moisture of the area.

The migration of radionuclides in the "soil-plant" system is the initial stage of the biological cycle, which subsequently can determine the level of radionuclide transfer to the organs and tissues of animals, as well as the products obtained from them.

The lowest transfer coefficients in soil-plant system ($T_{f\ (soil\text{-}plant)}$) of radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}Pu$, and ^{241}Am by plants were found at the sites of ground nuclear tests, increasing along the "trails" of radioactive fallout and in "background" areas. For zones of radioactive watercourses at the "Degelen" site and sites of testing combat radioactive materials ("4a" area), the highest accumulation coefficients have been established. The differences in K_acc for transuranic radionuclides $^{239+240}Pu$ and ^{241}Am do not exceed 15 times, for ^{137}Cs and ^{90}Sr – 75 times. The accumulation coefficients of radionuclides ^{137}Cs, ^{90}Sr,

$^{239+240}Pu$, and ^{241}Am by plants at different STS sites are presented by Larionova N et al. (2018, 2021).

2.5 Characteristics of Water Body Contamination at the STS

The STS in Kazakhstan encompasses several water bodies that have been contaminated due to nuclear testing activities. The radionuclide content in these water bodies varies significantly depending on their proximity to the sites of explosions and other factors.

Surface and Groundwater Characteristics

The surface water bodies within the STS are primarily comprised of the Shagan River and its tributaries. The area also features lakes and artificial water bodies of technogenic origin with varying levels of water mineralization, ranging from freshwaters to brines. Streams flowing from the Degelen mountain massif, including Uzynbulak, Karabulak, Baytles, and Tahtakushuk, contribute to the hydrological complexity of the region.

Groundwater in the STS is primarily sourced from atmospheric precipitation. The depth of groundwater is strongly correlated with the terrain's relief and soil cover. In intermountain areas with meadow, meadow-light chestnut soils, and meadow solonchaks, groundwater is found at depths of 2-6 meters. Beyond the Degelen mountain massif, under light-chestnut, shallow, and underdeveloped soils, groundwater levels drop to 10-12 meters, and in small hilly areas, it exceeds 10 meters.

Atomic Lake: Atomic Lake, formed by a nuclear explosion in 1965, exhibits notable contamination. The activity concentration of ^{137}Cs in the water of Atomic Lake is less than 0.01 Bq L^{-1} (Aidarkhanova et al., 2019). ^{241}Am levels are not available for this lake. ^{90}Sr in the water is measured at approximately 0.32 Bq L^{-1}. The concentrations of $^{239+240}Pu$ are very low, at around 0.0006 Bq L^{-1} (Aidarkhanova et al., 2018). ^{3}H, recorded at 140 Bq L^{-1} (Aktaev, M. R. et al., 2019).

Shagan Lake: Shagan Lake, another significant water body at STS, shows lower levels of radionuclide contamination compared to Atomic Lake. The activity of ^{137}Cs is less than 0.01 Bq L^{-1}. The levels of ^{241}Am are less than 0.03 Bq L^{-1}, while ^{90}Sr is recorded at less than 0.07 Bq L^{-1}. The concentration of $^{239+240}Pu$ in Shagan Lake is very low, at less than 0.001 Bq L^{-1}(Aidarkhanova et al., 2022). Tritium is measured at 40 Bq L^{-1} (Aidarkhanov A.O., et al. 2013, in Russian)

Shagan River: The Shagan River, the longest surface watercourse within the STS, flows through the eastern part of the Balapan site into "Atomic Lake," eventually forming a left-bank tributary of the Irtysh River. From 2016 to 2020, tritium levels in the Shagan River ranged from 8 Bq L^{-1} in the spring to 370,000 Bq L^{-1} in the summer-autumn period near "Atomic Lake." The primary source of contamination is groundwater leaching tritium into the river.

At the exit points of the Shagan River from the test site, tritium concentrations vary from 90 Bq L^{-1} to 12,400 Bq L^{-1}, and near the confluence with the Irtysh River, levels drop to below 110 Bq L^{-1}. The Shagan River, which flows through the contaminated area, has ^{137}Cs levels of less than 0.03 Bq L^{-1}. ^{241}Am data for the river is not available. The activity of ^{90}Sr is approximately 0.03 Bq L^{-1}. $^{239+240}Pu$ concentrations are less than 0.005 Bq L^{-1} (Aktayev M.R, 2021).

Crater No. 101: Crater No. 101, formed by an underground nuclear explosion, presents higher levels of contamination in comparison to other water bodies. The ^{137}Cs concentration in the water is about 0.10 Bq L^{-1}. The levels of ^{241}Am are less than 1 Bq L^{-1}. ^{90}Sr is recorded at approximately 0.023 Bq L^{-1}. The activity of $^{239+240}Pu$ is about 0.0011 Bq L^{-1}. Tritium levels are notably high, at around 310,000 Bq L^{-1} (Aidarkhanova et al., 2018).

The water from these sources is used by local farmers for agricultural purposes, including watering livestock. While some of these water bodies, like Shagan Lake, show relatively low levels of radionuclide contamination and might be considered safer for agricultural use, others, such as Shagan Riever, Atomic Lake and Crater No. 101, present significant risks due to higher radionuclide concentrations. Using water from highly contaminated sources can lead to the

accumulation of radionuclides in farm animals, affecting the safety of animal products such as milk and meat.

CHAPTER 3 MATERIALS AND METHODS OF RESEARCH

3.1 Research Location

The experiment was conducted on an Institute of Radiation Safety and Ecology (IRSE) Experimental Farm located on the STS territory.

The research took place at the "P-2" site within the "Experimental Field" area the STS (see Figure 1). The "Experimental Field" site occupies part of an inter-hill plain, representing a flat surface where above-ground and aerial nuclear explosions were previously conducted (1949–1962), thus forming the radioactive environment of the selected "P-2" site. These areas are among the most contaminated sites at the STS. The territory falls into the subzone of the deserted steppe with zonal light-chestnut soils under turf-grass vegetation. Moisturization occurs solely from atmospheric precipitation, which does not exceed 250 mm per year.

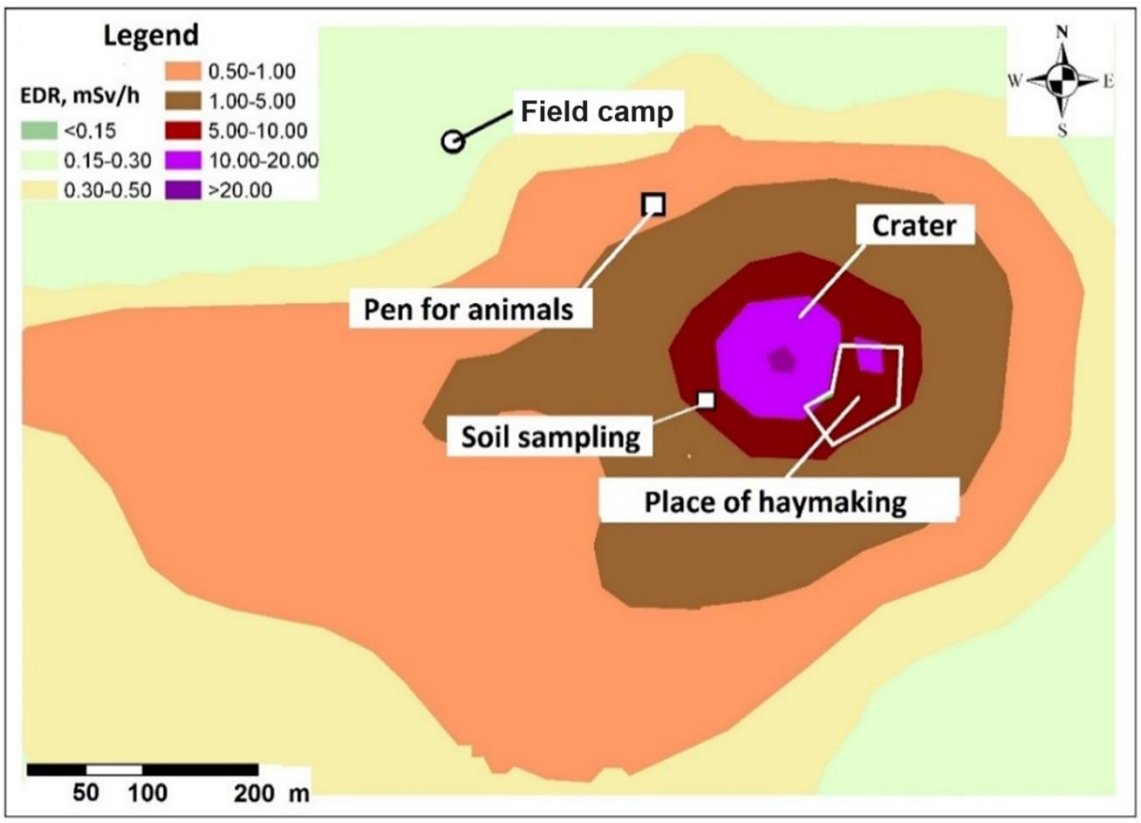


Figure 1.

Radiation characteristics of the hay and soil harvesting site

3.2 Research Subjects

The subjects of the research are the most common and typical agricultural animals (horses) and poultry for the region. Horses include typical regional non-breed (crossbreeds of Kazakh horses) broodmares aged 10–12 years and yearling fillies, with live weights of 350–400 kg and 150–200 kg, respectively. The type of agricultural poultry studied consists of 50±2 days old broilers (cross Arbor Acros). Animals and broilers in formed groups were similar in health status, exterior-constitutional data, and productivity. The radionuclides studied include ^{137}Cs, ^{90}Sr, ^{241}Am, and $^{239+240}Pu$.

3.3 Research Scheme for Studying the Transfer Parameters of ^{137}Cs, ^{90}Sr, ^{241}Am, and $^{239+240}Pu$ to Livestock Products

All studies were conducted during the summer in stable housing conditions for the animals. Separate groups of animals and broilers were fed contaminated feed, pre-prepared soil particles, given contaminated water, and in some cases, orally administered a water solution containing various radionuclides. The experiment was conducted at Institute of Radiation Safety and Ecology Experimental Farm (Baigazinov, 2019), which is located in the STS territory.

Before the experiment, all animals grazed on "clean" pastures with diverse steppe grasses. To adapt to new conditions after being transported to the research sites, animals and broilers were kept on a "clean" diet for 2 weeks.

The duration of animal and bird housing varied from 1 to 60 days. At the end of each term, following a 12-hour fasting period, the animals and broilers were slaughtered by bleeding. The slaughtering of farm animals was based on the rules approved by the Minister of Agriculture of the Republic of Kazakhstan (Order of the Minister of Agriculture of the Republic of Kazakhstan dated April 27, 2015 No. 7-1/370. It is registered in the Ministry of Justice of the Republic of Kazakhstan on July 8, 2015 No. 11591.).

Samples were taken for analysis: liver, kidneys, lungs, heart, spleen, thigh muscle, bone tissue, tongue muscles, brain. To monitor the intake of radionuclides into the bodies of animals and poultry throughout the experiment, daily records of the amount of consumed feed, soil, and drunk water were kept, as well as sampling of vegetation, soil, and water according to the experimental scheme. Subsequently, the collected and prepared biological samples were sent for spectrometric measurements.

3.3.1 Control samples

The animals were delivered from Sarzhal village (East-Kazakhstan) which is located more than 200 km to the south of the STS. Traditionally, farm animals in this region are allowed access to grazing all the year round.

Based on radioecological studies in the region of Sarzhal village, artificial radionuclides in soil are largely due to global fallout (Strilchuk, 2013). Analyses of farm products (meat, milk) from Sarzhal (Baigazinov, 2016) have shown that radionuclide activity concentrations were low (Table 1) so it was decided that a reference (or control) group was not required in our study. Previous research (Baigazinov Zh.A., 2010) has also established that the contribution of airborne radionuclides to the total radionuclide intake in animals and birds is minimal and can be effectively disregarded.

Table 1.

Activity concentration of the studied radionuclides in the farm products of Sarzhal, kBq kg^{-1} FM (Baigazinov, 2016)

Samples	^{241}Am	^{137}Cs	^{90}Sr	$^{239+240}Pu$
horse milk (n=3)	< 0.01	< 0.03	< 0.09	< 0.07
cow milk (n=7)	< 0.005	< 0.01	< 0.09	< 0.07
horse meat (n=2)	< 0.02	0.040±0.008	< 0.06	-

Note: FM – fresh mass (here and after)

3.3.2 Experiments with horses

We selected horses of cross Kazakh breeds which are typical of the region. The study group comprised two 10-12 year old pregnant mares and two 1-year old fillies with live-weights of about 350-400 and 150-200 kg respectively.

Study outline

The studies were performed during the summer when the mares and fillies were stabled. During the whole period of the experiment, all animals were in the same conditions regarding the duration of the adaptation period to new conditions, time in the stables, daily rations, feeding regime (2 times a day) and watering. The animals were fed 7-9 kg d^{-1} uncontaminated forage (steppe mixed grass hay –*Stipa capillata, Stipa sareptana, Festuca valesiaca, Artemisia sublessingiana, Artemisia frigida*) and 1.5 – 2 kg day^{-1} of wheat bran from a local flour mill. The daily consumption values are presented in dry weight. The mean concentration (±SD, n=24) of selected nutrient elements in the steppe mixed grass hay have previously been reported to be: Mg – 440±110 mg kg^{-1}, Ca – 290±70 mg kg^{-1}, *Sr* – 23±10 mg kg^{-1}, *Cs* – 0.03±0.01 mg kg^{-1} DM (Baigazinov, 2016). The concentration of K was not determined but from Kalashnikov (2003) it known that the concentration in the steppe mixed grass hay is about 500 mg kg^{-1}. Elemental concentrations in wheat bran had previously been reported to be: Mg – 4300 mg kg^{-1}, Ca – 2000 mg kg^{-1}, K – 11000 mg kg^{-1}, *Cs* – 0.03±0.01 mg kg^{-1} DM (Kalashnikov, 2003). Forage and water were delivered from outside the territory of the STS from Kurchatov city. Water was offered ad-libitum for all animals. Faeces and urine of animals were not collected in this experiment.

The animals were divided into two groups. Group 1 (one mare and one filly) was fed with contaminated soil mixed with wheat bran; the 2^{nd} group (one mare and one filly) was fed with wheat bran contaminated with a solution of mixed radionuclides extracted from contaminated soil (see below).

Contaminated soil-containing feed group (Group 1)

Preparation of the contaminated soil for feeding to the animals. Soil for feeding had been collected prior to the experiment from a site, where there have been surface nuclear explosions and which had high levels of radioactive contamination. The upper 5 cm of soil was sampled from the area of 8 – 10 m^2 and sieved through a 1.5-mm mesh. Sieved soil was placed in a container and thoroughly mixed.

The speciation of radionuclides in this soil have previously been determined (Kunduzbaeva, 2011, 2022); sequential extraction data from this earlier study are presented in the Table 2.

Table 2.

Form of radionuclides ^{137}Cs, ^{241}Am, $^{239+240}Pu$, ^{90}Sr in the study soil (Kunduzbaeva, 2011)

Speciation of radionuclides is soil	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
water-soluble(H_2O)	<0.01 %	<1.3 %	<0.4 %	<0.07 %
exchangeable(1M CH_3COONH_4)	<0.01 %	<1.2 %	<0.7 %	<1.4 %
organic(0.1N NaOH)	<0.3 %	<1.6 %	<0.4 %	<0.06 %
mobile(1M HCl)	1.0 %	14.0 %	<0.6 %	3.1 %
tightly bound	98.7 %	81.9 %	97.9 %	95.4 %

Note: Study of radionuclide speciation in soils was conducted by sequential extraction as modified by Pavlovskaya F.I. (1974). The procedure was modified by adding an intermediate stage of determining fractions of organically bound radionuclides by 0.1 NaOH solution based on the technique developed by Tyurin I.V (Ponomaryova, 1980). The ratio of soil and leaching solution was 1:5.

Feeding the animals with soil-containing feed. Every morning each animal was fed with a fresh mixture of 1 kg of soil and 2 kg of wheat bran. The animals consumed the offered feed in about 20 minutes. Observations of the feeding process confirmed that losses of the bran-soil feed mix due to spills or refused feed were minimal and comprised less than 1% of the total mixture volume. In the evenings, wheat bran (2 kg) was offered without any added soil.

Leachate -contaminated feed group (Group 2)

Preparation of radioactive solution for feeding the animals. Soil samples with high radionuclide content were taken from different areas of the STS: 1. surface nuclear explosion test site (mainly transuranic elements); 2. underground nuclear

explosion test site (mainly ^{137}Cs); 3. an area where radioactive substances had been dispersed for exploratory military purposes (mainly ^{90}Sr). Radionuclides were extracted from these soils for administration to the study animals.

Radionuclides were leached from the soils by boiling in an 8M solution of nitric acid for 2 hours. Radionuclides were then extracted from the resultant acidic leachate solution. Firstly, $^{239+240}Pu$ and ^{241}Am were separated by co-precipitation on metal hydroxides. Concentrated ammonia solution (pH 8-9) was then added to the acid solution. Hydrated oxides were filtered from the solution using a paper filter and then dissolved with a 2M hydrochloric acid solution. Secondly, ^{90}Sr was isolated by co-precipitating as carbonates. To achieve this 15-20 g of ammonium carbonate salt was added to the above filtrate and heated until it was completely precipitated. Strontium carbonate sediment was filtered and dissolved in a 2M hydrochloric acid solution. Finally, ^{137}Cs was isolated by co-precipitating using potassium hexacyanoferrate. Potassium hexacyanoferrate was added to the filtrate (from which ^{90}Sr had been removed) at a rate of 10 g of K- hexacyanoferrate per litre of filtrate and left for 24 hours. The resultant sediment was filtered and dissolved in the ammonia solution.

The resulting 9M hydrochloric acid solution contained $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr, the activity concentrations are presented in the Chapter 4.1.

Feeding animals with radioactively contaminated wheat bran. Every day during the experiment, 10 ml of the radioactive hydrochloric acid solution was added to 250 ml of distilled water and then neutralized with 60 ml of sodium hydroxide to achieve a solution pH of 2.5 (we found that precipitation started at pH 2.8). The solution was then added to 2 kg wheat bran and well mixed; the bran mixture was then offered to the animals. The animals consumed the offered feed in about 20 minutes. Observations of the feeding process confirmed that losses of the bran-solution feed mix due to spills or refused feed were minimal and comprised less than 1% of the total mixture volume. In the evenings, wheat bran (2 kg) was offered without any added solution.

3.3.3 Experiments with broilers

Study outline Part 1. Study of the accumulation of ^{241}Am and ^{137}Cs into the organs of the broilers

Objects of the study were 50±2 days old and 1700±100 g weight (live weight) broilers (cross *Arbor Acros*). They were purchased in a local poultry factory. All broilers were similar in clinical condition and external-constitutional data and they were kept in individual cages with a separate feeding and drinking bowl. During the experiment, the diet of all broilers was around 150-180 g/day/head (DW) commercial all-in-one granulated feed (hereinafter-all-in-one feed). Feeding was carried out twice a day in the morning (8:00-9:00 h) and evening (17:00-18:00 h). Before feeding, the all-in-one feed was steamed in warm water at a temperature of 20-25 ºC until the water was completely absorbed and the granules swelled (12-20 minutes). For all broilers water was offered ad-libitum.

The broilers were divided into two groups, each group had nine subgroups and each subgroup had three broilers. In total 54 broilers were used. During the whole period of the experiment, all broilers were in the same conditions (housing systems, watering and feeding regime) except for the source of radionuclide intake into the body. Group 1 was fed with contaminated soil mixed with the all-in-one feed; the 2nd group was fed with contaminated grass meal mixed with the all-in-one feed (see below). The duration of feeding with "contaminated" sources were 1, 2, 4, 8, 14, 28, 42, 56, 70 days (from each group one subgroup for each duration was used).

Group 1. Every morning each broiler was fed with a wet mixture of 30 g (DW) of soil and 50 g of all-in-one granulated feed. The broilers consumed the offered food in about 20-30 minutes. In the evenings, all-in-one granulated feed (100 g DW) was offered without any added soil.

Group 2. Every morning each broiler was fed with a wet mixture of 30 g (DW) of grass meal and 50 g of all-in-one granulated feed. The broilers consumed the offered food in about 30-40 minutes. In the evenings, all-in-one granulated feed (100 g DW) was offered without any added grass meal.

Soil for feeding was collected from a site of surface nuclear explosions test site called the "Experimental field" and which is known to have high levels of radioactive contamination. The test site territory fully described by Lukashenko (2020). Sampling was carried out at a distance of 20 meters from the funnel of one of the ground nuclear explosions. The upper 5 cm of soil was sampled from an area of 8 – 10 m^2 and sieved through a 0.5-mm mesh. Sieved soil was placed in a container and mixed up thoroughly. Afterwards the soil was packaged in plastic bags of 30 g each. In total, about 700 bags of soil were prepared before the start of the experiment. The speciation of radionuclides in this soil has previously been determined by Kunduzbaeva (2011, 2022) and sequential extraction data from this earlier study are presented in the Table 2 above.

Grass meal samples were made from vegetation taken from the test site territory called “Degelen”, which was designed for low-yield tests (up to several dozens of kilotons) in horizontal tunnels. At present, radionuclide contamination of the territory is caused due to the migration process of groundwater seepage through the epicenter (Lukashenko, 2020). At one of these sites, vegetation was mowed in mid of summer. The vegetation was presented meadow vegetation (*Chamaenerium angustifolium, Cirsium arvense, Tanacetum vulgare, Calamagrostis arundinacea, Urtica dioica, Veronica spuria, Mentha interrupta, Rumex confertus, Geranium collinum, Sanguisorba officinalis, Delphinium dictyocarpum etc.*). Mowed vegetation was washed to remove soil particles in the form of dust, dried to an air-dry state and ground to a state of grass meal (particle size 250 microns). Afterwards the grass meal was packaged in plastic bags of 30 g. In total, about 700 bags of grass meal were prepared before the experiment.

The daily intakes of radionuclides were estimated based upon activity concentrations (Bq kg^{-1}, DW) in the all-in-one feed, soil and grass meal and the daily consumption (kg day^{-1} DW) of each of them. Observations of the feeding process confirmed that losses of the feed-soil and feed-grass meal feed mix due to spills or refused feed were minimal and comprised less than 5% of the total mixture volume. No faecal matter or refused feed was analyzed.

The research did not perform a blank experiment, except three broilers' samples (muscle, liver, and bone) which were analyzed at the beginning of the experiment. It was found that activity concentration of radionuclides ^{137}Cs and ^{241}Am in the samples were below detectable activity.

Study outline Part 2. Study of the excretion of ^{241}Am and ^{137}Cs from the organs of the broilers

To study the excretion of ^{241}Am and ^{137}Cs, the observed animals (54 broilers) in the study were 50 ± 2 days old and 1700 ± 100 g weight (live weight) broilers (Cobb 500). They were purchased from a local poultry factory. All broilers were similar in clinical condition and external-constitutional parameters. They were placed in individual cages with a separate feeding and drinking equipment. During the experiment, the all broilers were fed around 150–180 g/day/head (DW) commercial all-in-one granulated feed (hereinafter-all-in-one feed). Feeding was carried out two times a day, once in the morning (8:00–9:00 h) and once in the evening (17:00–18:00 h). Before feeding, the all-in-one feed was steamed at a temperature of 20–25°C in warm water, until the water was completely absorbed and the granules swelled (12–20 min). For all broilers water was offered freely.

During the experiment, all broilers were kept in the same conditions, with the exception of the source of radionuclide intake into the body. The broilers were divided into two groups (27 broilers in each group). Group 1 was fed with all-in-one feed mixed with contaminated soil, while the 2nd group was fed with all-in-one feed mixed with contaminated grass meal. The duration of feeding with "contaminated" sources was 30 days. After 30 days, feeding continued with only all-in-one feed (not contaminated) until the end of the experiment. Then at 1, 2, 4, 8, 14, 28, 42, 56, 70 days one subgroup (3 broilers) from each group were exsanguinated based on the methods approved by the Minister of Agriculture of the Republic of Kazakhstan (2015).

Group 1. Every morning each broiler was given a wet mixture of 30 g (DW) of soil and 50 g of the all-in-one granulated feed for 30 days. The broilers consumed the offered food in about 20–30 min. In the evenings, all-in-one

granulated feed (100 g DW) was offered without any added soil. Then the feeding regime was changed to 150 g per day of the un-contaminated all-in-one granulated feed until termination, half in the morning, and half in the evening feeding period.

Group 2. Every morning each broiler was given a wet mixture of 30 g (DW) of grass meal and 50 g of all-in-one granulated feed for 30 days. The broilers consumed the offered food in about 30–40 min. In the evenings, all-in-one granulated feed (100 g DW) was offered without any added grass meal. Then the feeding regime was changed to 150 g per day of the uncontaminated all-in-one granulated feed until termination, half in the morning, and half in the evening feeding period.

Soil for feeding was collected from a surface nuclear explosion test site, which has high levels of radioactive contamination. The upper 5 cm of soil was collected from an area of 8–10 m^2 and sieved through a 0.5 mm mesh. Sieved soil was thoroughly mixed in a container. Afterwards the soil was divided into more than 800 pieces of 30 g plastic bags for the experiment. The radioactive characterization of radionuclides in this soil has been previously reported by Kunduzbaeva (2017);

Grass meal samples were prepared from vegetation taken from contaminated areas of the STS. The mowed vegetation was rinsed to remove soil particles in the form of dust, dried to an air-dry state and ground to grass meal (particle size 250 μm). Afterwards, the grass meal was divided into plastic bags of 30 g. More than 800 bags of grass meal were prepared for the experiment. Detailed information about soil and grass meal samples given in Mamyrbayeva et al. (2020).

The daily intakes of radionuclides were estimated based upon activity concentrations (Bq kg^{-1}, DW) in the all-in-one feed, soil and grass meal and the daily consumption (kg day^{-1} DW) of the boilers. Losses of feed-soil and feed-grass mixture due to spills and/or rejection were minimal and were less than 5% of the total amount, according to the observations of the feeding personnel. No fecal matter or refused feed was analyzed.

3.4 Analytical Work

3.4.1 Sampling of organs and tissues of horses

After 60 days of feeding the contaminated wheat bran, the animals were slaughtered by exsanguination. The slaughtering of farm animals was based on the rules approved by the Minister of Agriculture of the Republic of Kazakhstan (Order No. 11591, July 8, 2015). Organ sampling was conducted in accordance with established rules for sampling biological materials (Order No. 7-1/393, April 30, 2015 and Standardinform, 2011). On dissection both mares were found to be pregnant with foetuses aged approximately 3 – 5 months. The mass of the foetus in the soil group was 1.5 kg and that in the contaminated wheat bran group was 4.7 kg. Samples of muscle tissue (collected from the thigh), heart, liver, lungs, kidney and bones (ribs) were taken from each animal including the foetuses; tongue and spleen samples were also collected from the adults. From the foetuses gastrointestinal tract samples were collected; and carpal bones were also sampled from the Group 2 foetus.

Activities of $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr in muscle tissue, heart, liver, lungs, kidney and bones (ribs) were measured for the mares, fillies and foetuses. The radionuclide activities in spleen and tongue were determined for adult mares. In the foetuses, thigh bones and gastrointestinal tract contents were also analysed. Carpal bones were sampled from the foetus of the 2nd group.

Mare and filly samples mass comprised 1500 g; foetus ones – 150 and 200 g, for the first and the second foetus respectively. The total mass of mare's / filly's organs was: liver – 4700±100g / 3700±100g, lungs – 2300±100g / 1800±100g, heart – 1600±10g0 /1200±100g, kidneys – 850±50g / 650±50g spleen – 700±50g / 600±50g. Liver, lungs, heart, kidneys and spleen of the mares and fillies were divided into two parts: for radionuclide analyses, and preserve for further studies. The mass of the foetuses' liver, lungs, heart, kidneys were small (10–160 g) so the whole organs were used for analyses. Mare and filly ribs were separated from the meat and fat and then cut to 5-6 cm pieces. To get a rib mass sample of a foetus, we took 8-9 ribs making a 5 g mass for the first foetus and 9 g for the second

foetus. Complete femoral and carpal bones of the second foetus (20 and 10 g, respectively) were analysed; as was the femoral bone of the first foetus (6 g). Also, the gastrointestinal tract contents of the foetuses were analysed – the contents (30 g in the first foetus and 130 g in the second) were in the form of light-grey homogeneous liquid.

3.4.2 Sampling of organs and tissues of broilers

Samples of all-in-one feed, soil, grass meal and organs of all broilers (muscle, bone and liver) were taken for analysis. Three samples of all-in-one feed were taken before starting the feeding experiment from three different bags. Before Gamma-analysis, samples were ground by lab mill. Soil and grass meal samples were taken from the prepared feeding packages (5 packages each). Soil and grass meal samples preparation described above (see section 3.3.3).

The skin of the femoral muscle was removed, as well as the bone. Femoral bones from both legs were taken and cleaned from the muscle. At the beginning of experiment, the fresh weight of the liver, bone, and muscle samples were 34±5 g, 49±5 g and 4^{90}±40 g, respectively. The fresh weight of the liver, bone, and muscle samples of broiler from ninth subgroup (70^{th} day of the experiment) achieved 41±8 g, 82±11 g and 1030±140 g, respectively. Organs were prepared for analysis by washing with flowing water, drying (to constant weight) followed by grinding/homogenizing using a lab mill. The dry weight of the liver, bone, and muscle samples was 8.7±0.8 g, 20±3 g and 140±14 g at the beginning and 11±2 g, 49±6 g and 260±40 g at the end of experiment. The dry weight of the liver, bone, and muscle samples was 8.9±1.4 g, 21±2 g and 120±10 g, respectively. All samples were accurately weighed into plastic containers for gamma analysis depending upon sample size.

3.4.3 Preparation and measurement of animal samples for gamma and alpha-emitting radionuclides

Soft tissues were prepared for gamma-spectrometry by washing with flowing water and then grinding/homogenizing using a lab mill. Subsamples of 200g were taken for gamma-analyses. Bone samples were ashed at 380°C and the ash was then ground.

Gamma-spectrometric measurement of the samples under study was carried out using gamma spectrometers with Genie2000 software and high-purity germanium (HPGe) semiconductor detectors from MIRION (CANBERRA) and ORTEC. The laboratory was accredited according to ISO 17025:2009.

The evaluation of MDA during the measurements was assessed according to the formula:

$$MDA=\frac{4\sqrt{2}*\sqrt{n_f}}{\sqrt{t}*\varepsilon}$$

n_f – background count rate, cps (counts per second);
t – background accumulation time, s;
ε – sensitivity of the spectrometer to the given radionuclide (considering the spectrometer's detection efficiency and the gamma-ray emission intensity of the radionuclide).

Standard bulk reference sources containing ^{137}Cs, ^{241}Am, ^{152}Eu, ^{40}K, ^{238}U, and ^{232}Th and their decay products were used for the calibration of the spectrometer for detection efficiency (IAEA-RgTh, IAEA-RgU, IAEA-RgK Reference Materials-Archive (iaea.org), OMACH (special purpose volumetric measure containing ^{137}Cs, ^{241}Am, ^{152}Eu, a Russian source (Radium Institute named after V.G. Khlopin)).

The difference in self-absorption between the reference source and the measured sample was accounted for by the SpectraLine software package according to the expression:

$$\varepsilon_\rho(E) = \varepsilon_{\rho 0}(E)e^{(\mu\rho-\mu_0\rho_0)d}$$

μ, μ_0 – mass attenuation coefficients of the measured and reference sources;

ρ, ρ_0 – density of the measured and reference sources;

d – effective thickness of the measured sample.

Radiochemical separation and determination of ^{90}Sr and $^{239+240}Pu$

Following gamma-analyses samples were dried, charred on an electric stove and ashed in a muffle furnace at a 550°C until a white or light grey ash was obtained; the ashed samples were used for the subsequent determination of ^{90}Sr and $^{239+240}Pu$ activity concentrations. Ashed animal tissues were dissolved in conc. HNO_3 with the addition of H_2O_2 if required and ^{242}Pu was added as a yield tracer. The ^{90}Sr in the sample was determined from the daughter radionuclide - ^{90}Y. Subsequent to anion exchange and electrodeposition actinide activity concentrations were determined by alpha spectrometry. ^{90}Y measured on a liquid-scintillation beta spectrometer (Tri-Carb 2910 TR). The methodology used for $^{239+240}Pu$ and ^{90}Sr measurement has been described in a certified method (KZ.07.00.03614-2017).

The quantitative determination of $^{239+240}Pu$ in the samples was carried out by a radiochemical method using a tracer label – ^{242}Pu. The activity of $^{239+240}Pu$ was calculated using the tracer peak of ^{242}Pu in the energy range of 4850 – 4900 keV and the combined peak of ^{239}Pu and ^{240}Pu in the energy range of 5100 – 5170 keV obtained through alpha-spectrometric measurement of the counting sample.

When assessing the uncertainty of the activity, the uncertainties of the tracer peaks and $^{239+240}Pu$ were considered. The evaluation of MDA was carried out according to ISO 11929-7.

$$a^{\#} = \frac{2a^{*} + (k^2 w)/T}{1 - k^2 (\frac{u(w)}{w})^2}$$

$$a^{*} = \frac{k_{1-\alpha}}{m\varepsilon R}\sqrt{\frac{n_0}{Tt} + \frac{n_0}{tt}}$$

$$w = \frac{1}{m\varepsilon R}$$

$$k_{1-\alpha} = k = 1.65$$

a^* – detection threshold

n_0 – background count number

R – chemical yield

ε – detection efficiency

m – sample mass;

T – sample measurement time;

t – background measurement time.

The content of ^{90}Sr (by ^{90}Y) was estimated using the formula:

$$A = N_{cp}/(a \cdot b \cdot f_1 \cdot f_2 \cdot F)$$

N_{cp}. - is the average count rate of the sample, c^{-1};

a - is the correction factor for the chemical yield of yttrium, fraction;

b - is the correction factor for the chemical yield of strontium, fraction;

f_1 - is the coefficient accounting for the decay of ^{90}Y from the moment of separation from ^{90}Sr to the moment of beta radiation measurement;

f_2 - is the coefficient accounting for the accumulation of ^{90}Y in the ^{90}Sr solution until the moment of beta radiation measurement;

F – is the sensitivity of the radiometer.

3.4.4 Preparation and analyses of soil samples

Samples of 0.4-0.5 kg mass were air dried and the <1.5 mm fraction then separated by manual sieving. This fraction was quartered and a 200g sub-sample taken for gamma-analyses. After gamma-analyses the sample was finely ground using a pestle and a 10 g sub-sample was taken for further $^{239+240}Pu$ determination.

Then the 10-g mass were ashed at 450-500 °C and subjected to complete acid digestion. Extraction of plutonium isotopes was performed using column extraction chromatography followed by elution, co-precipitation and filtering through a membrane filter. Measurements of ^{137}Cs, ^{241}Am and $^{239+240}Pu$ activities were performed as described above for animal tissues.

After gamma-analysis a further 20 g soil sub-sampled was removed and placed in a measuring cuvette (d=70 mm). The entire cuvette volume was filled and the cuvette then well sealed and the ^{90}Sr activity concentration determined using a "Progress" beta-spectrometer. The ^{90}Sr determination did not use radiochemical separation, as expected activity concentration was in the order of kBq per kilogram. The minimum detectable activity concentration of ^{90}Sr was 100 Bq kg^{-1}. The ^{90}Sr determination method is fully described in an established method (KZ.07.00.00303, 2014).

3.4.5 Preparation and analyses of "uncontaminated" hay and wheat bran samples

The preparation of hay and wheat bran samples was carried out according to the method in GOST (2000). Gamma spectrometric analyses were carried out on a Canberra GX-2020 gamma spectrometer according to the standard method (MI 5.06.001.98). The radionuclide ^{90}Sr was determined by radiochemical isolation and measured on a TriCarb liquid-scintillation spectrometer according to an established method (KZ.07.00.00471-2005).

3.5. Estimation of transfer parameters

The standard parameters used to describe the transfer of radionuclides into farm animal tissues is the transfer coefficient (F_f; d kg^{-1}) (IAEA 2010):

$$F_f = \frac{\text{Radionuclide activity concentration in animal tissue (Bq kg}^{-1}\text{ fresh mass)}}{\text{Daily intake of a radionuclide (Bq d}^{-1}\text{)}}$$

Following the recommendations of Beresford et al. (2007b) and subsequently IAEA (2010) the dietary concentration ratio was also estimated where C_{Rdiet} is defined as:

$$C_{Rdiet} = \frac{\text{Radionuclide activity concentration in animal tissue (Bq kg}^{-1}\text{ fresh mass)}}{\text{Radionuclide activity concentration in the whole diet (Bq kg}^{-1}\text{ dry mass)}}$$

Daily intake of radionuclides for both groups were estimated based on the activity concentration of radionuclides in the contaminated soil and in the solution via their daily consumption with wheat bran and forage (soil – 1,000 g day^{-1}, solution – 10 ml day^{-1}, forage – 13 kg day^{-1} for mare and 11 kg day^{-1} for filly).

CHAPTER 4 TRANSFER OF RADIONUCLIDES TO LIVESTOCK AND POULTRY TISSUES

4.1 The transfer of $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr to the tissues of horses

4.1.1 Radionuclide intake by the study animals

Activity concentrations of $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr in the soil and leachate solution added to the diet of the study animals are presented in Tables 3 and 4. The estimated daily intakes of radionuclides were based upon the activity concentrations in the soil and leachate solution presented in Table 5.

Table 3.

Activity concentration of radionuclides in the soil fed to animals (Group 1)

	Activity concentration kBq kg^{-1} DM			
Sample	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
1	160±30	15±3	4.4±0.9	4.2±0.7
2	–	23±5	7.5±1.5	7.4±1.1
3	170±20	23±5	8.5±2.5	10.0±1.5
4	130±20	16±3	4.3±0.9	3.7±0.6
5	140±15	17±3	5.4±1.0	7.1±1.1
Mean ± SD	150±20	19±4	6±2	6.7±2.6

Note: Errors are analytical measurement errors, DM – dry mass, SD – standard definition

Table 4.

Activity concentration of 9M hydrochloric acid leachate solution, kBq l^{-1}

$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
18,800±900	922±50	45±2	1,500±150

Note: Errors are analytical measurement errors

Activity concentrations of ^{90}Sr, ^{137}Cs and ^{241}Am radionuclides in all samples of offered forage (mixed grass hay) and wheat bran were <2 Bq kg^{-1}, <1 Bq kg^{-1} and <0.1 Bq kg^{-1}, respectively. No measurement of $^{239+240}Pu$ in the feed were performed.

Table 5.

The daily dietary intake (kBq day^{-1} DM) and radionuclide activity concentration in the whole diet (kBq kg^{-1} DM)

Group	Radionuclides (Mean ± SD)							
	$^{239+240}Pu$		^{241}Am		^{137}Cs		^{90}Sr	
	mare	filly	mare	filly	mare	filly	mare	filly
Daily intake of radionuclides, kBq day^{-1} DM								
1st group	150±20		19±4		6±2		6.7±2.6	
2nd group	188±9		9.2±0.5		0.45±0.02		15±2	
Radionuclide activity concentration in the whole diet, kBq kg^{-1} DM								
1st group	10.7±1.3	12.5±1.6	1.4±0.3	1.6±0.3	0.44±0.15	0.52±0.17	0.48±0.18	0.56±0.21
2nd group	14.5±0.7	17.1±0.9	0.71±0.04	0.84±0.04	0.035±0.002	0.041±0.002	0.56±0.07	0.48±0.06

Note: The errors - are the of SD of mean activity concentration of radionuclides in soil or leachate solution

However, based upon the measured ^{241}Am activity concentrations we would expect $^{239+240}Pu$ activity concentrations to be <1 Bq kg^{-1}. Based on this, the intake of artificial radionuclides with uncontaminated feed was negligible. This evidence also supported the decision that a control group was not necessary, as the potential influence of artificial radionuclides in the uncontaminated feed was minimal

4.1.2 Radionuclide activity concentrations in horse tissues

Radionuclide activity concentrations in animal tissues are summarised in Table 6. There were relatively more samples from Group 1 with activity concentrations below detection limits compared to Group 2. The Group 1 foetus had most measured values below the detection limit, however, this foetus only had about 30% of the mass of the Group 2 foetus. Activity concentrations of ^{137}Cs in the tissues of Group 2 animals were typically about two orders of magnitude higher than those determined in tissues from Group 1 animals, even though the daily intake of ^{137}Cs was about ten times higher for Group 1 compared with Group 2 animals. Similarly, whilst the ^{90}Sr intake of Group 2 animals was higher than that of Group 1 animals, ^{90}Sr activity concentrations in Group 2 animal tissues were disproportionately higher than those of animals in Group 1.

Table 6.

Activity concentration of $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr in organs and tissues of the mares and the fetuses, Bq kg^{-1} FM

Tissue	Group 1 (source of radionuclides - soil)			Group 2 (source of radionuclides - solution)		
	mare	filly	foetus	mare	filly	foetus
			$^{239+240}Pu$			
muscle	n.a.	0.021±0.003	0.34±0.03	0.140±0.004	0.11±0.01	0.22±0.02
liver	1.2±0.1	3.6±0.1	0.018±0.01	59.7±1.5	41.5±1.3	0.32±0.03
lungs	0.15±0.01	0.33±0.02	0.025±0.01	2.7±0.08	1.8±0.1	0.034±0.01
heart	0.016±0.005	0.083±0.01	<0.06	0.51±0.01	0.32±0.1	0.095±0.030
kidneys	0.19±0.01	1.20±0.05	0.029±0.020	7.6±0.2	1.1±0.1	0.088±0.020
spleen	0.19±0.02	0.052±0.007	n.a.	4.8±0.1	2.8±0.1	n.a.
ribs	0.32±0.07	1.2±0.1	2.7±0.4	9.3±0.4	20.1±2.0	2.8±0.4
femoral bones	n.a.	n.a.	0.28±0.20	n.a.	n.a.	1.8±0.2
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	2.4±0.4
gastrointestinal tract content	n.a.	n.a.	0.10±0.04	n.a.	n.a.	0.310±0.030
			^{241}Am			
muscle	0.022±0.008	< 0.03	< 0.04	0.06±0.02	< 0.03	< 0.04
liver	n.a.	0.45±0.04	< 0.09	6.6±1.0	5.5±1.1	< 0.05
lungs	0.08±0.03	0.15±0.03	< 0.09	0.33±0.04	0.36±0.03	< 0.09
heart	0.09±0.04	0.11±0.04	<0.09	0.08±0.03	0.15±0.04	< 0.1
kidneys	< 0.08	0.23±0.04	< 0.09	1.21±0.08	3.52±0.30	< 0.1
spleen	0.19±0.07	0.13±0.06	n.a.	0.49±0.06	0.28±0.07	n.a.
ribs	0.24±0.1	< 0.26	4.4±2.0	1.3±0.2	2.11±0.4	< 1.5
femoral bones	n.a.	n.a.	< 3.0	n.a.	n.a.	< 1.3
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	<0.5
gastrointestinal tract content	n.a.	n.a.	< 0.2	n.a.	n.a.	<0.1
			^{137}Cs			
muscle	0.23±0.02	0.79±0.06	0.13±0.05	12.1±0.2	22.2±0.2	3.0±0.1
liver	0.27±0.10	0.75±0.09	< 0.2	5.2±2.0	11.8±3.0	1.1±0.1
lungs	< 0.1	< 0.2	< 0.2	3.8±0.2	7.6±0.2	2.1±0.2
heart	0.40±0.1	0.53±0.1	n.a.	13.2±0.3	17.7±0.4	2.2±0.3
kidneys	< 0.2	< 0.2	< 0.2	3.9±0.2	6.2±0.3	3.2±0.3
spleen	< 0.2	< 0.2	n.a.	9.4±0.3	14.1±0.5	n.a.
ribs	< 0.1	< 0.2	n.a.	2.6±0.3	4.9±0.7	3.3±1.0
femoral bones	n.a.	n.a.	n.a.	n.a.	n.a.	2.4±0.9
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	<3.3
gastrointestinal tract content	n.a.	n.a.	< 0.45	n.a.	n.a.	3.1±0.2
			^{90}Sr			
muscle	<3.0	0.42±0.2	< 1	10±1	10±1	15±1
liver	3.4±0.1	1.6±0.1	< 2	24±1	14±1	13±1
lungs	3.5±0.2	2.0±0.2	2.4±1.0	54±1	35±1	10±1
heart	0.52±0.2	1.4±0.6	<7	19±1	13±1	11±2
kidneys	3.3±0.3	6.4±0.3	< 2	120±1	564±2	6±1
spleen	2.1±0.5	2.1±0.4	n.a.	18±1	12±1	n.a.
ribs	900±13	3,330±20	87±10	2,860±20	10,830±30	380±10
femoral bones	n.a.	n.a.	103±11	n.a.	n.a.	7,450±30
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	870±20
gastrointestinal tract content	n.a.	n.a.	< 3	n.a.	n.a.	2,000±20

Tissue	Group 1 (source of radionuclides - soil)			Group 2 (source of radionuclides - solution)		
	mare	filly	foetus	mare	filly	foetus

The errors are analytical measurement errors
Note: "n.a." – not available; "<" – Activity concentrations below the minimum detectable activity (MDA).

The relative activity concentrations in the different tissues of the animals were generally similar to the values described in the literature for all studied isotopes (e.g. see Yankovich et al. 2010; Coughtrey et al. 1984 a,b)

The highest activity concentrations of $^{239+240}Pu$ in the mares and fillies were found in liver and the lowest observed in muscle. These data are in good agreement with the findings of previous studies (e.g. Buldakov et al. 1969; Howard & Lindley 1985; Beresford et al. 2007a, Fesenko et al. 2009a). For the foetuses, $^{239+240}Pu$ activity concentrations were highest in bone; this is in agreement with the findings of the studies reviewed by Coughtrey et al. (1984a). We have to acknowledge that the activity concentrations determined in the tissues of our study animals after feeding contaminated diets for 60 days will not be at equilibrium as the biological half-life of *Pu* is in the order of many years (Durbin, 1975; Fesenko et al. 2015; Gilbert et al. 1989). Such long biological half-lives have led to the suggestion that *Pu* activity concentrations are unlikely to ever reach equilibrium over the lifetime of productive agricultural or wild mammals (Howard et al. 2007; Johansen et al. 2016).

Similarly to the $^{239+240}Pu$ activity concentrations, ^{241}Am values are highest in the liver of the mares and fillies with the exception of the Group 1 mare. The comparatively high concentrations in liver are in agreement with previous observations on other mammal species (Coughtrey et al. 1984b; Beresford et al. 2007a). However, differences between the tissue activity concentrations were less than those observed for $^{239+240}Pu$. For the Group 1 foetus ^{241}Am activity concentrations were, similar to $^{239+240}Pu$ highest in bone. Biological half-life values for *Am* are in the order of many years, so the $^{239+240}Pu$, ^{241}Am activity concentrations in the study animals would not have reached equilibrium with concentrations present in the diet (Coughtrey et al. 1984b; Gilbert et al. 1989).

As observed previously (e.g. see Yankovich et al. 2010) ^{137}Cs was relatively evenly distributed in the various soft tissues of the mares, fillies and foetuses. The highest radionuclide activity concentrations of this radionuclide have been found in heart and muscle tissues of the animals; the lowest, in bones. Activity concentrations in the tissues of the foetus were lower than in those of the mare with the exception of rib bone for Group 2 (no foetus bone data being available from Group 1).

The highest ^{90}Sr activity concentrations in mare and fillies were found in bones as expected for a calcium analogue. Bone ^{90}Sr activity concentrations were higher in the fillies than the mares which is as would be expected for still developing animals with a higher calcium demand (NRC 1989). Bone ^{90}Sr concentrations in the Group 2 foetus were considerably higher than those of the Group 1 foetus. The lower mass of the Group 1 foetus suggests that this animal was less developed (younger) than the Group 2 foetus. This observation is in agreement with Skuterud et al. (2005), who report that the transfer of Ca and *Sr* to the foetus increases in late pregnancy.

4.1.3 Radionuclide transfer parameters

Estimates values of transfer coefficient and concentration ratio for the four radionuclides considered are presented in Tables 7 and 8 respectively. For all radionuclides transfer parameters are higher for Group 2 (radionuclide intake source = leachate solution) than Group 1 (radionuclide intake source = soil). Soils from the STS contain radioactive particles and that these particles have comparatively high activity concentrations of *Pu* (IAEA, 2011) and also contain *Cs*, *Sr* and *Am*. The particles have low solubility for instance, Salbu et al. (2018) state that the extraction of ^{241}Am from soils collected in the STS using 0.16 M HCl was ‘very low’, 72-85% of the activity remained unextrated from the sample even after 168 hours. Radionuclides associated with such particles will be largely unavailable for adsorption from the gastrointestinal tract; similarly, the transfer of ^{137}Cs and *Pu* from ingested soils has been shown to be low compared to

radionuclides ingested in 'soluble' forms (Beresford et al. 2000). Therefore, the lower transfer parameters derived for animals ingesting contaminated soil from the STS is expected. However, contaminated soil is realistically likely to dominate the intake of most radionuclides by animals grazing in the STS and given the low plant uptake of *Pu*, soil ingestion is likely to dominate the intake of *Pu* by farm animals in this case, despite its lower transfer parameters. This highlights the need to incorporate the soil-product or soil-animal direct pathway in addition to the soil-vegetation-product pathway when assessing the radiological exposure from or the feasibility of economical use of heavily contaminated areas.

Table 7.

Estimated transfer coefficients (F_f) for $^{239+240}Pu$. ^{241}Am. ^{137}Cs and ^{90}Sr from soil and radioactive solution to organs and tissues of the mares and the fetuses. d kg^{-1} FW

Organs	Group 1 (source of radionuclides - soil)			Group 2 (source of radionuclides - solution)		
	mare	filly	foetus	mare	filly	foetus
$^{239+240}Pu$. n×10^{-5}						
femoral muscles	n.a.	0.014±0.003	0.23±0.05	0.074±0.020	0.059±0.02	0.12±0.05
liver	0.8±0.2	2.4±0.6	0.012±0.003	31.8±8	22±6	0.17±0.05
lungs	0.10±0.03	0.22±0.05	0.017±0.01	1.4±0.4	0.96±0.30	0.018±0.006
heart	0.011±0.003	0.055±0.02	<0.04	0.27±0.08	0.17±0.05	0.051±0.02
kidneys	0.13±0.03	0.8±0.2	0.019±0.010	4.0±1.0	0.59±0.20	0.047±0.01
spleen	0.13±0.03	0.035±0.01	n.a.	2.6±0.8	1.5±0.5	n.a.
ribs	0.21±0.05	0.8±0.2	1.8±0.5	4.9±1.0	10.7±3.0	1.5±0.4
femoral bones	n.a.	n.a.	0.19±0.05	n.a.	n.a.	0.96±0.3
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	1.3±0.3
gastrointestinal tract content	n.a.	n.a.	0.067±0.02	n.a.	n.a.	0.16±0.05
^{241}Am. n×10^{-5}						
femoral muscles. lungs. heart. spleen[a]	0.45 (0.34-0.61)	0.68 (0.63-0.74)	<0.5	2.2 (0.82-4.0)	3.0 (2.3-3.5)	<0.6
liver	n.a.	2.4±0.6	<0.5	72±22	60±18	<0.6
kidneys	<0.4	1.2±0.3	<0.7	13±4	38±11	< 0.6
ribs	1.3±0.4	<1.4	23±7	14±4	23±7	<16
femoral and carpal bones	n.a.	n.a.	<15	n.a.	n.a.	<14
gastrointestinal tract content	n.a.	n.a.	<1	n.a.	n.a.	<1
^{137}Cs. n×10^{-3}						
All organs[a]	0.045 (0.042-0.056)	0.13 (0.11-0.13)	<0.022	11.6 (8.6-23.9)	26.2 (15.3-35.3)	6.0 (4.8-6.9)
^{90}Sr. n×10^{-3}						
femoral muscles. liver. lungs. heart. spleen. [a]	0.41 (0.25-0.51)	0.24 (0.21-0.30)	<0.4	1.2 (1.2-1.6)	0.88 (0.81-0.91)	0.77 (0.69-0.87)
kidneys	0.49±0.19	0.96±0.38	<0.4	8.0±1.6	38±8	0.41±0.08

Organs	Group 1 (source of radionuclides - soil)			Group 2 (source of radionuclides - solution)		
	mare	filly	foetus	mare	filly	foetus
ribs	134±54	500±200	13±5	190±38	720±144	25±5
femoral bones	n.a.	n.a.	15±6	n.a.	n.a.	500±100
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	58±11
gastrointestinal tract content	n.a.	n.a.	<0.4	n.a.	n.a.	130±27

Note: [a] – Median (lower and upper quartile); The errors are reflective of differences of analytical measurement errors and SD of daily radionuclide activity consumed; "n.a." - not available. "<" – estimated F_f values (based on the minimum detectable activity)

Table 8.

Estimated concentration ratio (C_R) of the radionuclide activity concentration in the tissues (Bq kg^{-1} FW) of the mares and the fetuses to the activity concentration of the total diet of the animals

Organs and tissues	Group 1 (source of radionuclides - soil)			Group 2 (source of radionuclides - solution)		
	mare	filly	foetus	mare	filly	foetus
	$^{239+240}Pu$. n×10^{-6}					
muscle tissue	n.a.	1.7±0.4	32±7	10±3	6.4±2	15±6
liver	112±28	290±72	1.7±0.4	4130±1100	2430±700	22±8
lungs	14±4	26±7	2.3±0.6	190±50	105±30	2.4±0.7
heart	1.5±0.4	6.6±1.7	<5.6	35±11	19±6	6.6±2
kidneys	18±5	96±24	2.7±0.7	530±130	64±20	6.1±2
spleen	18±5	4.2±1.1	n.a.	330±100	160±50	n.a.
ribs	30±8	96±25	250±80	640±190	1170±350	194±70
femoral bones	n.a.	n.a.	26±7	n.a.	n.a.	125±40
carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	166±50
gastrointestinal tract content	n.a.	n.a.	9.3±3	n.a.	n.a.	21±6
	^{241}Am. n×10^{-5}					
femoral muscles. lungs. heart. spleen [a]	6.3 (4.8-8.5)	8.2 (7.6-8.8)	<7	29 (11-52)	33 (26-38)	<13
liver	n.a.	28±8	<7	930±300	660±200	<8
kidneys	<6	15±5	<7	170±50	420±130	<17
ribs	18±5	<16	320±100	180±50	250±80	<210
femoral and carpal bones	n.a.	n.a.	<200	n.a.	n.a.	<180
gastrointestinal tract content	n.a.	n.a.	<15	n.a.	n.a.	<14
	^{137}Cs. n×10^{-4}					
muscle tissue [a]	6.3 (5.8-7.8)	15.0 (12.8-15.4)	<6.8	1500 (1110-3110)	2880 (16^{90}-38^{90})	780 (630-^{90}0)
	^{90}Sr. n×10^{-3}					
femoral muscles. liver. lungs. heart. spleen. [a]	6.3 (4.4-7.1)	2.9 (2.5-3.6)	<4	16 (15-20)	10 (9-10)	10 (9-11)
kidneys	6.9±2.8	11.5±4.6	<4	104±25	410±80	5.3
ribs	1880±700	5960±2400	180±72	2480±500	7940±1600	330±80
femoral bones	n.a.	n.a.	215±90	n.a.	n.a.	6460±1600

carpal bones	n.a.	n.a.	n.a.	n.a.	n.a.	750±190
gastrointestinal tract content	n.a.	n.a.	<6	n.a.	n.a.	1730±400

Note: [a] – Median (lower and upper quartile); The errors are reflective of differences of analytical measurement errors and SD of daily radionuclide activity consumed; "n.a." - not available. "<" – estimated C_R values

As discussed above, activity concentrations of ^{241}Am and $^{239+240}Pu$ in the study animals will not have reached equilibrium. However, there are few studies reporting transfer coefficient values for actinide elements and the same criticism could justifiably be made about any of them. The IAEA handbook of transfer parameters (IAEA, 2010) gives recommended values for *Pu* for beef and mutton (rams meat) of $1.1x10^{-6}$ d kg^{-1} and $5.3x10^{-5}$ d kg^{-1} (with an overall range of $8.8x10^{-8}$ to $3.0x10^{-4}$ d kg^{-1}) respectively; values for horsemeat (muscle) in Table 7 are within the reported range for other livestock. There are only single ^{241}Am F_f values quoted for both beef and mutton in IAEA (2010) both of which are in the 10^{-4} d kg^{-1} range, higher than values for horsemeat determined in this study. The transfer coefficient values for ^{241}Am in mutton after soil ingestion ($2.3\ x10^{-6}$ d kg^{-1}) (Baigazinov, 2016) are in the same order of magnitude with the horse values of Group 1 ($1.2\pm0.4\ x10^{-6}$ d kg^{-1}).

The large difference in F_f values for ^{137}Cs determine for Group 1 compared to Group 2 animals is in agreement with other studies, which have shown that the transfer of *Cs* ingested as soil can be up to about 50 times lower than that for *Cs* administered as *Cs*Cl or incorporated into vegetation by root uptake (Beresford et al. 2000). The muscle F_f values determined for ^{137}Cs for the mare and filly in Group 2 (leachate solution) are higher than values for beef cited in IAEA (2010) and more similar to those cited for rams and goats. The estimate of F_f for Group 2 animals is about an order of magnitude higher than that reported for a Kazakh breed horse administered of $3.5x10^{-2}$ d kg^{-1} ^{137}Cs via water for 90 days (Semioshkina et al. 2006). There are a number of differences between the two studies, including stage of pregnancy (early stage in the current study, late stage and after foaling in the experiment by Semioshkina et al. (2006), milk production, duration of the study, way and amount of intake, which could all have influenced the results. Furthermore, both experiments were conducted on a very small number

of animals, order of magnitude differences can be observed in the IAEA (2010) handbook for different animals as well.

The ^{90}Sr muscle F_f values for the Group 2 mare and filly are similar to the recommended values for beef, mutton and pork in IAEA (2010) and also the value determined for a horse administered ^{90}Sr via water ($3.0x10^{-3}$ d kg^{-1}) for ^{90} days (Semioshkina et al. 2006). The transfer of *Sr* to animals is largely determined by its' Ca status, the animals' dietary intake and demand for Ca (Beresford et al. 1998); unfortunately we do not have data on the Ca concentrations in the diet of our study horses. Beresford et al. reported that the Ca status of the animals was the controlling factor determining transfer of *Sr* from soil contaminated feed across the gastrointestinal tract, not the dietary source (Beresford et al. 2000). The lower *Sr* transfer of *Sr* associated with ingested soil observed in the study reported here may be the consequence of *Sr* being associated with radioactive particles.

We have also quantified the transfer of radionuclides to tissue of the study animals as the concentration ratio (CR_{diet}) for the soil group the feed mass include the mass of soil. The difference in between Group 1 and Group 2 are the same as that for F_f (compare Tables 7 and 8). Comparing CR values for other animals' meat (beef, mutton, and pork) in IAEA (2010) with Group 2 showed that they are similar, whereas CR for Group 2 has big differences. For instance, mean CR for *Cs* in beef, mutton, pork is 2.3×10^{-1}, 6.4×10^{-1}, 9.2×10^{-2} respectively (IAEA, 2010) while, for mare's meat it is 3.5×10^{-1} for Group 2 and 5.2×10^{-4} for Group 1. CR for *Am* for mutton is 1.1×10^{-4} (IAEA, 2010) for mares' meat 8.5×10^{-5} for Group 2 and 1.6×10^{-5} for Group 1; for *Pu* it is the same as for *Am*. While the values for horse are similar to those of other ruminants and monogastrics in IAEA 2010, there are three orders of magnitude differences between Group 1 and Group 2, indicating the importance of feed source. The use of conservative single values has some merit in time critical applications, however if the time and resources are available, a case specific investigation might be warranted.

4.1.4 Conclusions for Chapter 4.1

This research presents transfer parameter data for $^{239+240}Pu$, ^{241}Am, ^{137}Cs and ^{90}Sr to horsemeat from contaminated soil and feed, which is an important agricultural product in the STS. One of the study diets involved the feeding of contaminated soil collected from within the STS. The transfer of radionuclides from ingested soil was generally lower, by up to three orders of magnitude, than from a diet contaminated by a leachate solution. The ingestion of soil is a particularly important source of radionuclide intake to grazing animals in the STS (Beresford and Howard, 1991; Hinton et al., 1995).

Activity concentrations of ^{241}Am and $^{239+240}Pu$ in the study animals will not have reached equilibrium during the duration of the study.

For ^{241}Am there is a lack of available data, the two singular entries for mutton and beef in the IAEA handbook are higher than all values observed in the current study. The maximum observed transfer coefficient was $72\pm22\times10^{-5}$ d kg^{-1} FW in the liver of the Group 2 mare fed with leachate contaminated feed. In general, Group 2, with the leachate contaminated feed had had higher transfer values compared to Group 1, which had soil contaminated feed.

For $^{239+240}Pu$ the maximum transfer coefficient was $31.8\pm8\times10^{-5}$ d kg^{-1} FW observed also in the liver of the Group 2 mare. The values are within the range of values reported for beef and mutton by the IAEA (IAEA, 2010). In general, Group 2, with the leachate contaminated feed had had higher transfer values compared to Group 1, which had soil contaminated feed.

The filly from Group 2 had the highest transfer parameter values for ^{137}Cs, an average of 26.2×10^{-3} d kg^{-1} FW, with the range of 15.3×10^{-3} d kg^{-1} FW to 35.3×10^{-3} d kg^{-1} FW. The muscle F_f values determined for ^{137}Cs for the mare and filly in Group 2 (leachate solution) are higher than values for beef cited in IAEA (2010) and more similar to those cited for rams and goats. The Group 2 animals had transfer coefficients about an order of magnitude higher than that reported for a Kazakh breed horse previously (Semioshkina et al. 2006). Group 1 had significantly lower transfer coefficients (more than two orders of magnitude)

confirming previous studies reporting a significantly lower availability of *Cs* from soil compared to contaminated feed or water (Beresford et al. 2000).

The highest ^{90}Sr transfer coefficient was found in the ribs of the filly from Group 2, 720±144 $\times 10^{-3}$ d kg^{-1} FW. The ^{90}Sr muscle F_f values for the Group 2 mare and filly are similar to the recommended values for beef, mutton and pork in IAEA (2010) and also the value determined for a horse administered ^{90}Sr via water (3.0×10^{-3} d kg^{-1}) for ^{90} days (Semioshkina et al. 2006), while Group 1 fed soil contaminated feed had somewhat lower values.

The results presented in this research are based on a low sample size; future studies need to use a larger number of animals. However, this study has provided data for a poorly studied farm animal and, in the case of ^{241}Am and $^{239+240}Pu$, relatively poorly studied radionuclides with respect to transfer to animal products.

4.2 The Dynamics of Accumulation and Transfer Parameters of ^{241}Am and ^{137}Cs to Broiler Tissues

4.2.1 Radionuclide intake by the study animals

The live-weight of the broilers increased from 1700±100 g to 3300±200 g over the 70 days of the study; The mean (±SD) dry matter intakes of the study Group 1 and Group 2 broilers during the study was 180±10 g/day.

Activity concentrations of radionuclides measured in soil and grass meal samples are presented in Table 9. Each one of the sources (soil, grass meal) contains kBq per kg of dry matter level of contamination. The SD value of the mean activity concentration of radionuclides in soil and grass meal were not more that 10% and it is assumed that the activity concentration of radionuclides in the prepared sources was uniform in the total volume. As it was expected the activity concentrations of radionuclides (^{137}Cs and ^{241}Am) in the all-in-one feed was below the minimum detectable activity. Intakes of radionuclides by the study broilers, taking into account the mass of refused feed (not more than 5% of each source), are summarized in Table 10.

Table 9.

Mean radionuclide activity concentration in soil and grass meal offered to the broilers during the experiment, Bq kg^{-1} DM

Sample Number #	Soil (Group 1)		Sample Number #	Grass meal (Group 2)	
	^{137}Cs	^{241}Am		^{137}Cs	^{241}Am
1	1 600±100	340 000±30 000	1	5 800±300	620±30
2	1 400±100	430 000±40 000	2	5 700±300	550±30
3	1 600±100	420 000±40 000	3	5 500±300	570±30
4	1 500±100	440 000±40 000	4	5 500±300	640±30
5	1 500±100	430 000±40 000	5	5 700±300	660±30
Mean±SD	1 500±200	410 000±60 000	Mean±SD	5 600±300	610±60

Table 10.

Radionuclide activity concentration in the whole diet and daily intake of radionuclides

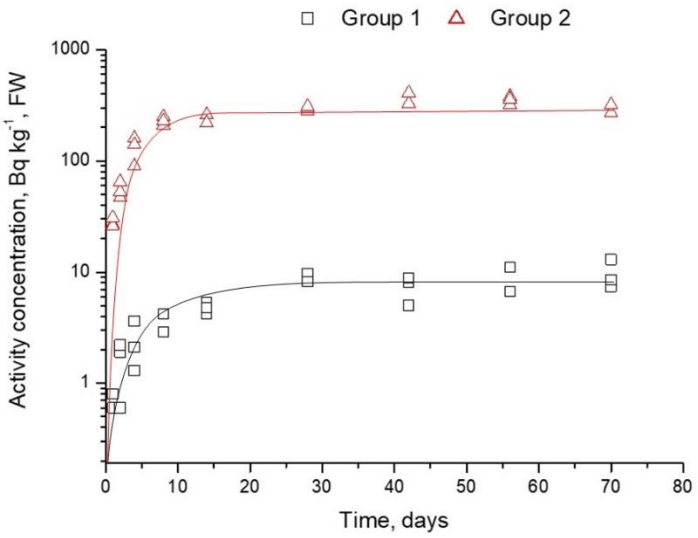
Group	Radionuclides (Mean±SD)	
	^{137}Cs	^{241}Am
Daily intake of radionuclides a, Bq day^{-1} DM		
1st group	46±8	12 000±1 500
2nd group	170±30	18±3
Radionuclide activity concentration in the whole diet, kBq kg^{-1} DM		
1st group	250±50	66 700±12 000
2nd group	940±200	5 300±500

Note: a The SD associated with the daily activity reflective of differences in dry matter intake between broilers and SD of mean activity concentration of radionuclides in soil and grass meal

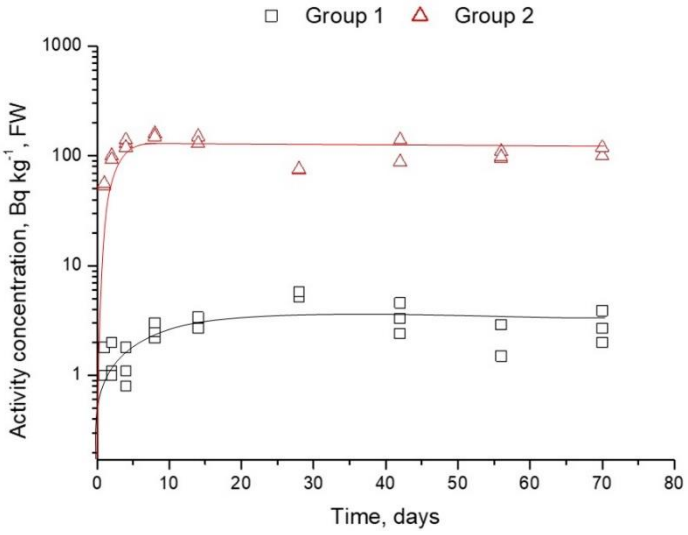
Figure 2 and 3 present the fresh weight (FW) radionuclide mean (±SD) activity concentration of ^{137}Cs in the muscle, liver and bone of Group 1 and 2. ^{137}Cs was detectable in almost all analyzed samples. Activity concentration of ^{137}Cs in the muscle of broilers in Group 1 and 2 in last day of experiment reached 6-13 Bq kg^{-1} FW and 300-400 Bq kg^{-1} FW, respectively. At the same, time the activity concentration of ^{137}Cs in the muscle is higher 2-3 and 5-6 times than in liver and bone, respectively. The relative activity concentrations in the different tissues of the broilers were generally as would be expected from the literature (e.g. see Yankovich et al. 2010; Coughtrey et al. 1984 a,b)

Unfortunately, the activity concentrations of ^{241}Am in Group 2 were below the minimum detectable activity, but in Group 1 ^{241}Am was detectable in almost all analyzed samples (Figure 3); as expected ^{241}Am mainly deposited in liver and bone, muscle activity concentration is much lower than in others. Comparatively high concentrations in liver and bone are in agreement with previous observations on mammal species (Coughtrey et al. 1984b; Beresford et al. 2007a)

a)



b)



c)

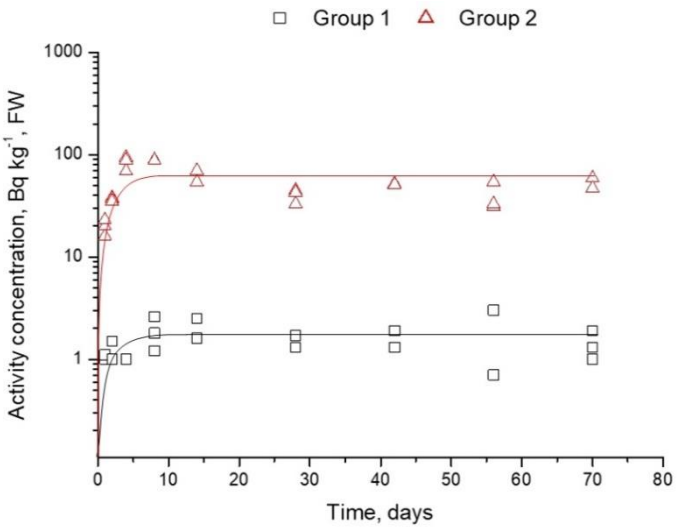


Figure 2.

Mean (±SD) activity concentrations of ^{137}Cs in the muscle (a), liver (b) and bone (c) during long term intake, Bq kg^{-1}, FW (The analytical error of measurements no more than 10%)

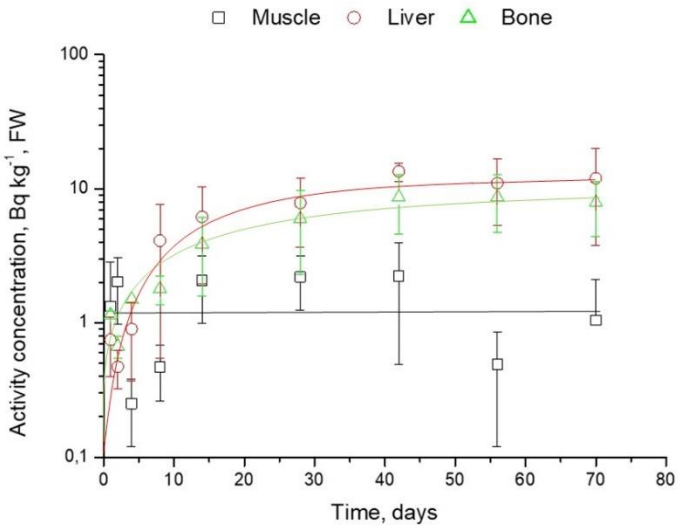


Figure 3.

Mean (±SD) activity concentrations of ^{241}Am in the organs of broilers Group 1 (n=27) during long term intake, Bq kg^{-1}, FW

4.2.2 Dynamics of Accumulation

Figure 2 has shown the activity concentration of ^{137}Cs in organs increases to a certain value, after which accumulation of radionuclides slow down, i.e. dynamic equilibrium occurs between accumulation and elimination. For bone and liver, it has come after 14 days, for muscle after 30 days. These data are in good agreement with the findings of previous studies (Shilov and Koldayeva, 1978) after ^{137}Cs was administered as a chloride into the body. However, Sirotkin (1987) showed that transfer coefficient of ^{137}Cs of chicken meat for 30 days feeding was 2.4 times lower than with 390 days feeding. Astasheva (1991) reported that biological half-life accumulation of adult geese muscles was 5 days, and for liver it was 1 day.

Compared to ^{137}Cs, the deposition of ^{241}Am in organs is different. Activity concentrations of ^{241}Am in muscle are almost the same in the first and last day of the experiment, while in liver and bone it increased 5-10 times. Previously, it was determined that *Am* in farm (Ham, 1989, Gilbert, 1989) and laboratory animals (Green, 1977, Humphreys, 1982, Priest, 1977) accumulates in the liver and skeleton. The dynamics of transuranic elements' metabolism in the liver can be explained by the dynamics of the exchange of complex compounds of the radionuclide with globulins (Netchev, 1977). The figure 3 above shows that,

similarly to ^{137}Cs in muscle, ^{241}Am in the bone swiftly increases in the first 40 days of the experiment, after that the growth of radionuclide activity slows down. However, in case of ^{241}Am in the liver, it is not clear if it is exactly at the equilibrium stage, but it is obviously slowing down. In addition, it should be mentioned that the range of ^{241}Am activity concentrations in organs in the same subgroups (n=3) could be two times different, while for instance the range of ^{137}Cs is less than 20%.

4.2.3 Transfer coefficient (F_f) and Concentration ratio (C_R)

F_f and C_R were calculated for the equilibrium stage of radionuclides into the organs. For ^{137}Cs in muscle equilibrium stage was considered after a duration of feeding more than 28 days (4 subgroups in each group), for liver and bone more than 14 days (5 subgroups in each group); for ^{241}Am in liver and bone-more than 42 days (3 subgroups), for muscle-from first day of intake (all 9 subgroups). Table 11 shows that difference between Group 1 and 2 of F_f of ^{137}Cs in broilers organs could reach one orders of magnitude while for C_R it reaches three orders of magnitude. F_f and C_R values to muscle tissue are higher than they are to the liver, regardless of the source of intake, by 2.5-3.5 times.

Table 11.

Transfer coefficient and Concentration ratio of ^{137}Cs in broilers' organs, mean±SD, 10^{-2}

Organs	N	Transfer coefficient			Concentration ratio		
		Group 1	Group 2	IAEA (2010)	Group 1	Group 2	IAEA (2010)
Muscle	11	18.8±4.6	192.9±25.8	270.0* (120–560)	0.073±0.018	34.7±4.7	39.0**
Liver	14	7.8±3.1	57.6±12.4	-	0.031±0.012	10.4±2.2	-
Bone	14	3.4±1.3	26.4±5.7	-	0.013±0.0052	4.7±1.0	-

Note: *-Mean and range of F_f values for *Cs* in poultry meat which summarized 13 data including data for duck older than 40 days and duration of feeding with contaminated fed, not less than 20 days'; **-generated values of C_R for all animals' meat (beef, rams and pork)

Comparing with summarized F_f values in the IAEA handbook (2010) the obtained data for Group 2 is lower than the mean data, at the same time it is within the range of the data presented in the handbook. C_R for meat of broilers Group 2

(Table 11) is comparable with values generated for the meat of different animals (IAEA, 2010). It was found that F_f and C_R data that obtained for Group 1 is lower one and two orders of magnitude respectively, than it is other experiments where soil was used as a source of intake (*Am*aral, 1995).

F_f of ^{241}Am into broiler meat is lower by three orders of magnitude than it is for ^{137}Cs. As the range of ^{241}Am in muscle in equilibrium stage was high, it is recommended to use median data (Table 12). Our data review (IAEA, 2010; Fesenko, 2009; Green, 2003; Yankovich, 2009) showed that transfer data for ^{241}Am into chicken meat was not available yet. In comparison, the generated C_R values of ^{241}Am for meat of different animals (IAEA, 2010) is one order of magnitude higher than the obtained C_R data for Group 1. At the same time these differences for ^{137}Cs could reach three orders of magnitude (Table 11).

Table 12.

Transfer coefficient and Concentration ratio of ^{241}Am in broilers of group 1, $\times 10^{-4}$

Organs	N	Transfer coefficient		Concentration ratio		IAEA (2010)
		Mean±SD	Median ($Q_{1/2} - Q_{3/4}$)	Mean±SD	Median ($Q_{1/2} - Q_{3/4}$)	
Muscle	27	1.1±0.95	0.75 (1.4-0.33)	0.20±0.18	0.14 (0.28-0.06)	1.1*
Liver	6	10.1±4.5	10.0 (12.5-7.04)	1.8±0.82	1.8 (2.3-1.3)	-
Bone	9	7.03±2.8	9.2 (9.2-3.4)	1.3±0.50	1.7 (1.7-0.62)	

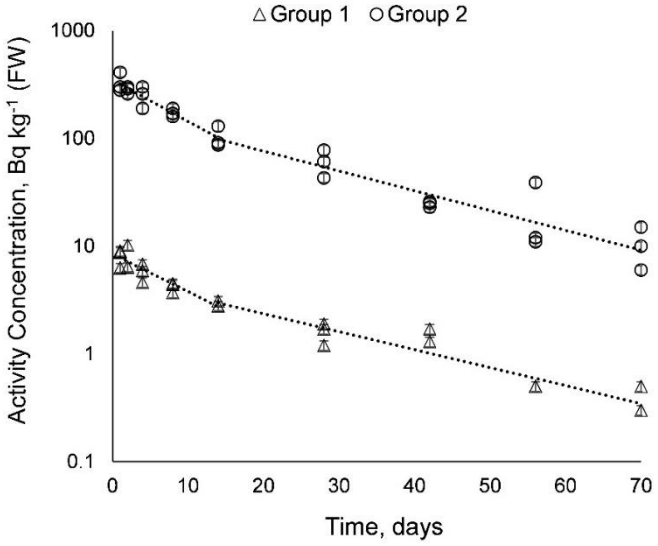
Note: *-generated values of C_R for all animals meat

4.2.4 The Dynamics of Excretion of ^{241}Am and ^{137}Cs from the Broilers Organs After Long-Term Application

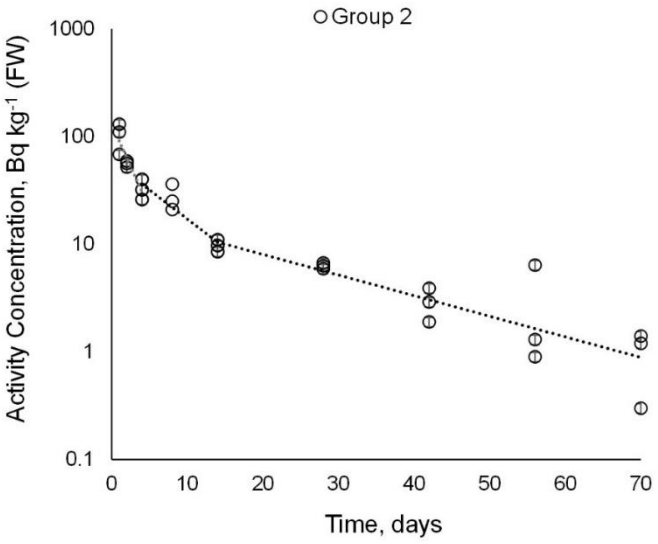
Radionuclide intake by the study animals described in the chapter 4.2.1

Figures 3 (a, b, and c) present the fresh weight (FW) radionuclide activity concentration of ^{137}Cs in the muscle, liver, and bone for Group 1 and 2 for the period from 1 up to 70 days after 30-day application. ^{137}Cs was detectable in all analyzed samples in Group 2 and in only in muscle samples in Group 1. The activity concentrations of ^{137}Cs in other samples (liver, bone) in Group 1 were detected only on the first day after 30-days application.

a) muscle



b) liver



c) bone

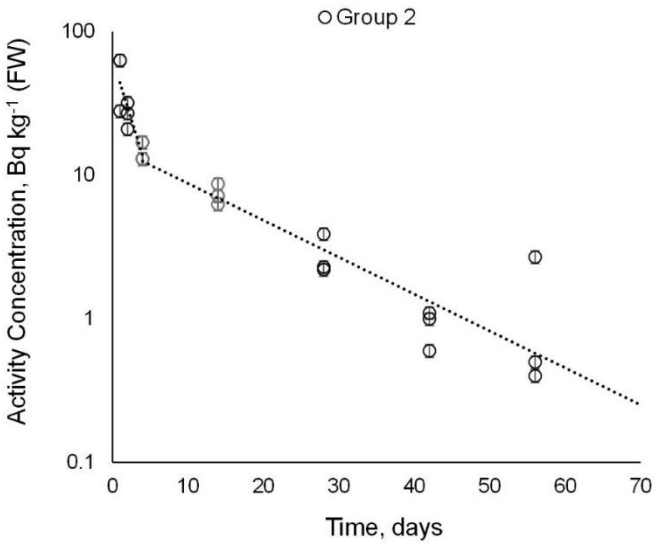


Figure 4.

Dynamics of excretion of ^{137}Cs in the muscle (a), liver (b) and bone (c) of Group 1 and 2 broilers 30 days after application, Bq kg^{-1}, FW

Activity concentrations of ^{137}Cs in the muscle of broilers in Group 1 and 2 on the first day after long term application were 6-9 Bq kg^{-1} FW and 300-400 Bq kg^{-1} FW, respectively. The activity concentrations of ^{137}Cs in the muscle are higher 2-3 and 5-6 times than in liver and bone, respectively. On the 70^{th} day, only 2-8% of the first-day activity concentrations of ^{137}Cs in organs (muscle, liver, bone) were detected. The relative activity concentrations in the different tissues of the broilers were generally as would be expected from the literature (e.g. see Yankovich et al. 2010; Coughtrey et al. 1984 a,b).

The dynamics of the excretion of ^{137}Cs in muscle (Fig. 4), liver, and bone could be described by exponential decrease. The bulk of the radionuclide was excreted from the organs in the first week. The analysis of the curves showed that the curves could be divided into several components: a fast and a slow excretion period, which can be approximated by equations (1-7):

$$\mathbf{A}_{\mathbf{c\ muscle}}^{\mathbf{(t=1\ldots14)}} = 274\ e^{-(0.084\pm0.004)\ t} \qquad (1)$$

$$\mathbf{A}_{\mathbf{c\ muscle}}^{\mathbf{(t=14\ldots70)}} = 177\ e^{-(0.040\pm0.002)\ t} \qquad (2)$$

$$\mathbf{A}_{\mathbf{c\ liver}}^{\mathbf{(t=1\ldots4)}} = 130\ e^{-(0.36\pm0.02)\ t} \qquad (3)$$

$$\mathbf{A}_{\mathbf{c\ liver}}^{\mathbf{(t=4\ldots14)}} = 59\ e^{-(0.12\pm0.01)\ t} \qquad (4)$$

$$\mathbf{A}_{\mathbf{c\ liver}}^{\mathbf{(t=14\ldots70)}} = 19\ e^{-(0.044\pm0.004)\ t} \qquad (5)$$

$$\mathbf{A}_{\mathbf{c\ bone}}^{\mathbf{(t=1\ldots4)}} = 65\ e^{-(0.39\pm0.02)\ t} \qquad (6)$$

$$\mathbf{A}_{\mathbf{c\ bone}}^{\mathbf{(t=4\ldots70)}} = 15\ e^{-(0.059\pm0.003)\ t} \qquad (7)$$

Caesium largely behaves similarly as potassium and reaches ubiquitously all tissues especially those with high Na/K ATP-ase activity such as muscles, heart and kidneys. Generally, the dynamics of the excretion of ^{137}Cs in the different organs of the broilers is in accordance with the values reported by Ng et al. (1982);

Andersson et al. (1990), Voigt et al. (1993), Amaral et al. (1995), Pöschl et al. (1997), Shilov et al. (1978).

The activity concentrations of ^{241}Am almost in all analyzed samples of Group 2 were below the minimum detectable activity (Table 13), but in Group 1 ^{241}Am was detected in almost all organs.

Table 13.

Activity concentrations of ^{241}Am in the broiler's organ 30 days after application, Bq kg^{-1}, FW

Time after 30-days application, days	Muscle			Liver			Bone		
Group 1									
1	0.7±0.1	0.3±0.1	1.1±0.2	4.4±0.9	12±2	12±2	5.1±0.5	10.4±0.6	11.2±0.7
2	2.8±0.6	BDA	n.a.	9.7±1.9	1.9±0.4	n.a.	4.1±0.5	2.5±0.4	n.a.
4	4.5±0.9	1.6±0.3	0.14±0.03	1.6±0.3	7.0±1.4	3.5±0.7	2.5±0.5	4.5±0.5	3.2±0.5
8	BDA	0.3±0.1	0.7±0.1	2.8±0.6	1.1±0.2	2.3±0.5	1.9±0.5	2.2±0.4	3.5±0.5
14	0.9±0.2	0.15±0.03	0.13±0.03	16±3	1.1±0.2	7.4±1.5	16.2±0.9	2.5±0.4	10.5±0.6
28	BDA	0.7±0.14	0.13±0.03	2.3±0.5	3.4±0.7	1.7±0.3	4.3±0.5	3.6±0.4	1.9±0.4
42	0.12±0.02	0.22±0.04	BDA	5.1±1.0	34±7	2.7±0.5	3.6±0.5	n.a.	3.0±0.4
56	0.13±0.03	BDA	BDA	1.2±0.2	1.4±0.3	1.7±0.3	5.2±0.5	3.5±0.4	2.7±0.3
70	BDA	2.5±0.5	BDA	2.4±0.5	0.7±0.1	1.6±0.3	5.5±0.5	1.3±0.3	2.7±0.4
Group 2									
1	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
2	0.4±0.1	0.5±0.1	2.6±0.5	BDA	BDA	BDA	BDA	BDA	2.2±0.4
4	BDA	BDA	0.4±0.1	BDA	BDA	BDA	BDA	BDA	BDA
8	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
14	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
28	n.a.	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
42	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
56	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA
70	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA	BDA

Note: BDA – below detectable activity (0.1 Bq kg^{-1}), n.a.- not available.

As expected, ^{241}Am mainly metabolized in liver and bone, activity concentration of ^{241}Am in muscle is much lower. This data good agreement with previous observations (Coughtrey et al. 1984b; Beresford et al. 2007a). However, despite the fact that muscle tissue is not the main organ of ^{241}Am deposition, we can observe its presence up to the last day of the experiment. Similar results were obtained by Rysina (1962) and Belyaev (1962) in the spleen (rabbits, dogs), which

is not also the site of the actinides deposition, 3-4 months after the introduction of radionuclides.

In addition, as is shown in Table 3, the activity concentrations of ^{241}Am in each subgroup (n=3) have a quite wide range. For bone and liver samples it could be up to 2-3 time difference, but for the muscle it could reach up to 10 times. The wide range of the activity concentrations of ^{241}Am in organs could be explained by the individual differences between animals, however all other conditions such as animal welfare, age, weight, feed source, etc. were almost identical.

As it has been already noted, currently there is not much information on the transfer parameters of transuranic radionuclides to livestock products, with the exception of some results on the transfer coefficient and distribution of radionuclides in farm animal organs (e.g. Averin. et al. 2011; Howard et al. 2007, 2009; Beresford et al. 2007a; Green & Woodman, 2000; Yankovich, 2010; Fesenko, 2015). The kinetics of metabolism, microdistribution, and excretion of the radionuclide from the body experiments were carried out mainly only on small laboratory animals and a common way of radionuclide application was injection. We acknowledge that there is the possibility of injected radionuclides to behave differently from ingested radionuclides within an animal (e.g. see Mayes et al., 1996), but we assume that these data are comparable and provide some basic knowledge about this process.

As it is shown in Table 3, the activity concentration of ^{241}Am in the muscle, liver, and bone of broilers in Group 1 on the first day after the 30-day application were 0.3-1.0, 4-12, and 5-11 Bq kg^{-1} FW, respectively. After 70 days, only 20-50% of the first-day activity concentrations of ^{241}Am in the liver were detected. The activity concentration of ^{241}Am in the muscle on the 70th day of the experiment was below the detectable activity. On the 56th day, approximately 15% of first-day activity concentrations were detected.

Fig. 4 shows that the excretion of ^{241}Am from muscle, liver, and bone has a different curve, which can largely depend on the metabolic activity of different organs or tissues (Korneyev and Sirotkin, 1987). Over the first two days the

activity concentration of ^{241}Am decreases twofold in both the liver and bone. After that, we can see that the activity concentration in the bone sample is kept constant until the last day of the experiment at around 35% of the maximum activity concentration. The activity concentration in the liver slowly decreases and 15% of the maximum activity concentration remained to the last day of the experiment.

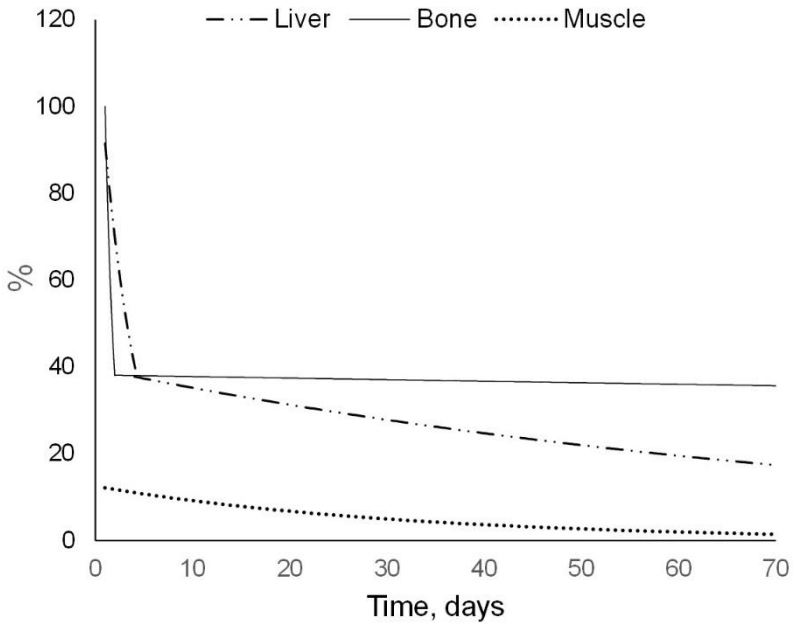


Figure 5.

Dynamics of excretion of ^{241}Am in the muscle, liver and bone of broilers 30 days after application, percent of maximum activity concentration

The curves (Fig. 5) of excretion of ^{241}Am in the muscle, liver and bone of broilers 30 days after application can be approximated by equations from (8) to (12).

$$\mathbf{A}_{\mathbf{c\ muscle}}^{\mathbf{(t=1\ldots70)}} = 12\ e^{-(0{,}031\pm0{,}003)\ t} \qquad (8)$$

$$\mathbf{A}_{\mathbf{c\ liver}}^{\mathbf{(t=1\ldots4)}} = 120\ e^{-(0{,}27\pm0{,}02)\ t} \qquad (9)$$

$$\mathbf{A}_{\mathbf{c\ liver}}^{\mathbf{(t=4\ldots14)}} = 40\ e^{-(0{,}012\pm0{,}001)\ t} \qquad (10)$$

$$\mathbf{A}_{\mathbf{c\ bone}}^{\mathbf{(t=1\ldots2)}} = 263\ e^{-(0{,}97\pm0{,}15)\ t} \qquad (11)$$

$$\mathbf{A}_{\mathbf{c\ bone}}^{\mathbf{(t=2\ldots70)}} = 38\ e^{-(0{,}0008\pm0{,}0002)\ t} \qquad (12)$$

Biochemical processes behind the distribution of transuranium elements are very sparsely reported or theoretized in the literature. Mostly mammals were investigated in all previous reports. Therefore, any such biochemical process investigation would need to be derived from a specially designed tracing experiment specially in birds. Plus, there are no known physiological mechanisms that facilitate the uptake of any actinide from the alimentary tract (Ménérier et al. 2003).

Dynamics of excretion of ^{241}Am in the organs of broilers were explained via analogies. Americium exists mainly in its Me(III) oxidation state. Its absorption from the alimentary tract is very poor, in line with that of all actinides`. Once absorbed from the epithelial cells of the small intestine, to the blood, it might bind to transferrin as other trivalent cations. Transferrin might bind and shuttle the ions of any trivalent cation to liver and bone marrow, where it might enter an enterohepatic cycle and also be able to bind to the mineral compartment of bone through electrochemical interactions. Before reaching this „slow" release curve, ^{241}Am kidney excretion might be responsible for the „fast" exponential part of the curve.

It was found that unlike the liver, ^{241}Am is firmly fixed in the bone tissue, the activity concentration is kept constant throughout the entire observation period (see Fig. 2). The same result could be observed in Stepanov's experiment (1970) regarding excretion of ^{241}Am from the rats' liver and bone. Also, the same dynamics of excretion were found in other studies with rat, mice, dogs, and rabbits (Buladkov 1969, 1979; Isimov, 1978; Rysina, 1962; and Belyaev, 1962), but with some differences in time of excretion of radionuclides from organs.

4.2.5 Conclusion for Chapter 4.2

This research presents transfer parameter data for ^{241}Am, ^{137}Cs to broilers' meat, which is an important agricultural product for the inhabitants living nearby the STS. The study diets involved the feeding with feed contaminated with soil and vegetation collected from within the STS. The transfer of radionuclides from ingested soil is generally lower, by up to one order of magnitude, than that from a

diet including contaminated grass meal. The ingestion of soil and grass are a relevant source of radionuclide intake for feeding broilers on contaminated territory (Beresford and Howard, 1991). Our study has provided data for poorly studied poultry meat, and in case of ^{241}Am, a relatively poorly studied radionuclide with respect to transfer to animal products.

The obtained results confirm the previously available data on the dynamics of the excretion of ^{137}Cs from organs, which can be described with a fast and a slow exponential curve excretion. On the 70th day, only 2-8% of the first-day activity concentrations of ^{137}Cs in organs (muscle, liver, bone) were detected. Excretion of ^{241}Am from broilers' liver and bone is slower than that of ^{137}Cs. After 70 days, 20-50% of the first-day activity concentrations of ^{241}Am in liver were detected. Activity concentration of ^{241}Am in the muscle on the 70th day of experiment were below the detectable activity, on the 56th day, approximately 15% of first-day activity concentrations were detected. ^{241}Am is practically not removed from the bone, the activity concentration is kept fairly constant throughout the entire observation period. Our study has provided data for poorly studied poultry meat, and in case of ^{241}Am, a relatively poorly studied radionuclide with respect to excretion from broilers' tissues.

CHAPTER 5 ASSESSMENT OF THE POSSIBILITY OF LIVESTOCK FARMING ON THE STS

5.1 Regulatory Framework of the Republic of Kazakhstan on the Quality Assessment of Livestock Products

In the Republic of Kazakhstan, the regulatory framework for assessing the quality of livestock products is specifically designed to address the challenges posed by historical nuclear testing in the Semipalatinsk region. On April 8, 2022, the Minister of Energy of the Republic of Kazakhstan, B. Akchulakov, approved the "Methodology for Conducting Comprehensive Environmental Assessments of Territories Where Nuclear Weapons Testing Was Conducted" for conducting comprehensive environmental assessments of areas affected by nuclear testing. This document outlines procedures for evaluating radiation levels in soil, water, air, and biota, and assessing potential radiation exposure to local populations. The goal is to identify radiation hazards and recommend appropriate mitigation measures. Key regulatory documents include Annex 6, which provides a methodology for calculating radionuclide intake in livestock products.

Annex 6 details the process for estimating radionuclide intake in livestock products when animals graze on contaminated land. The methodology includes sampling and analysis of soil, water, and forage to determine radionuclide concentrations. It calculates radionuclide intake based on animal diet and grazing patterns, applies T_f to estimate radionuclide levels in animal tissues and products, and compares these levels with safety standards.

This regulatory framework aims to protect both the environment and public health by ensuring that livestock products from areas affected by nuclear testing meet stringent safety standards.

The calculation of the activity concentration of radionuclides in livestock products (meat, milk, eggs) is carried out according to the formula:

$$A_{i,\ product} = V_{forage} \times A_{i,forage} \times T_{f,forage}$$

where:

$A_{i,\ product}$ – activity concentration of the i-th radionuclide in livestock products (milk, meat, eggs), Bq kg^{-1};

$V_{\ foraged}$ – daily forage intake, kg/day;

$A_{i,\ forage}$ – activity concentration of the i-th radionuclide in the forage, Bq kg^{-1}. The activity concentration of radionuclides in the forage is taken as the activity concentration of radionuclides in the feces of hoofed animals. In the absence of data on the activity concentration of radionuclides in the feces of hoofed animals, the maximum value of the activity concentration of radionuclides in the vegetation cover in each designated zone is taken as the activity concentration of radionuclides in the forage according to point 38 of the Methodology (ME RK, 2022);

$T_{f\ forage}$ – Transfer Coefficient of the radionuclide from the forage to 1 (one) kilogram or liter of the product.

The transfer coefficients of radionuclides from the forage to livestock products (meat, milk, eggs) are calculated according to the formula:

$$T_{f\ forage} = A_{i,\ product} / A_{i,\ forage}$$

5.2 Maximum Permissible Levels of Radioactive Contamination of Food

The Hygienic Standards for Ensuring Radiation Safety, approved by the Order of the Minister of Health of the Republic of Kazakhstan on August 2, 2022, No. RK DCM-71 (MH RK 2022), define the permissible levels of ^{137}Cs and ^{90}Sr in food products. For livestock products, the following activity concentration limits have been established:

- For meat, meat products, and offal, the permissible levels are set at 200 Bq kg^{-1} for ^{137}Cs and 50 Bq kg^{-1} for ^{90}Sr.
- For poultry and poultry by-products, the limits are 180 Bq kg^{-1} for ^{137}Cs and 80 Bq kg^{-1} for ^{90}Sr.
- For milk (raw materials) and cream (raw materials), the permissible levels are 100 Bq kg^{-1} for ^{137}Cs and 25 Bq kg^{-1} for ^{90}Sr.

The content of radionuclides $^{239+240}$Pu and ^{241}Am in food products is not standardized in the Hygienic Standards of Kazakhstan. Therefore, the permitted levels established by the European Union are utilized. The European Parliament (2017) has set maximum permissible levels of radioactive contamination for various foodstuffs in the event of a nuclear accident or radiological emergency. Specific limits for alpha-emitting isotopes of plutonium and transplutonium elements, including ^{239}Pu and ^{241}Am, are as follows:

- Dairy produce: 20 Bq kg^{-1}
- Other food (except minor food): 80 Bq kg^{-1}
- Liquid food: 20 Bq kg^{-1}

It is important to note that the permissible levels for ^{137}Cs in dairy produce in Kazakhstan are ten times lower, and in other food products (excluding minor food) are six times lower than those established in the European Union regulations.

5.3 Agricultural Activities and Monitoring at the STS

During the monitoring of economic activities within the STS, various agricultural entities engaged in farming activities were identified. Typically, these are winter settlements where agricultural animals are kept. Mostly settlements are located in the southeastern part of the STS. Information about these winter settlements was updated through site visits and interviews with their residents.

In total, approximately 140 winter settlements were registered within the STS. These settlements are sparsely populated, with the number of residents ranging from 2 to 8 people per settlement, except for the village of "Samay," where 70 people reside. The inhabitants are predominantly ethnic Kazakhs. The diet of the residents mainly consists of products from their own livestock, such as meat and milk. The structures in these settlements are predominantly made of adobe.

Water for drinking and watering livestock is sourced from wells located in close proximity to the winter settlements. The residents' diet mainly consists of products from their own livestock, with occasional purchases of flour, grains, and canned goods from nearby trading points.

The primary activities in these winter settlements are livestock farming (sheep, cattle, and horse breeding) and haymaking. Livestock grazing is carried out on lands adjacent to the settlements, with grazing radii for cattle up to 5 km, small ruminants up to 10 km, and horses up to 15-25 km. As grazing is conducted freely (without constant supervision), grazing areas can expand. Free grazing of animals has been repeatedly observed around the perimeter of the Degelen mountain massif and within the valleys of its streams or on the "4a" site.

The primary livestock activities in the territory of the STS and its adjacent areas include the breeding of small ruminants, cattle, camels, and horses. Livestock products, such as goat meat, cow and goat milk, as well as chicken meat and eggs, are primarily used to meet the farmers' personal needs. Mutton, beef, horse meat, and mare's milk are produced for commercial sale in nearby cities and districts.

Currently, within the surveyed territory of the test site, which covers 40% of the total area of the STS, 42 active farms have been identified. These farms support over 250 people engaged in livestock activities and house more than 2,500 heads of cattle, 13,000 heads of small ruminants, and 1,600 heads of horses. The livestock breeds in the STS area include coarse-wooled fat-tailed rams, crossbreeds of meat and dairy cattle, and Kazakh breed horses such as Jabe and Abaev.

Livestock is maintained under stable-pasture conditions, with year-round grazing conducted using a free-range method. Some farms, however, restrict pasturing to the summer period only. This system supports the efficient utilization of available forage resources while sustaining the local agricultural economy.

Figure 6 presents a schematic map of the STS with the marked locations of farm enterprises and testing areas.

When assessing the quality of livestock products, the meat and milk used for food were sourced from residents of farms located within the STS. These products, obtained directly from the STS territory, were subsequently subjected to spectrometric measurements, the detailed methodologies for which are described in Chapter 3. The results of these measurements are presented in Table 14.

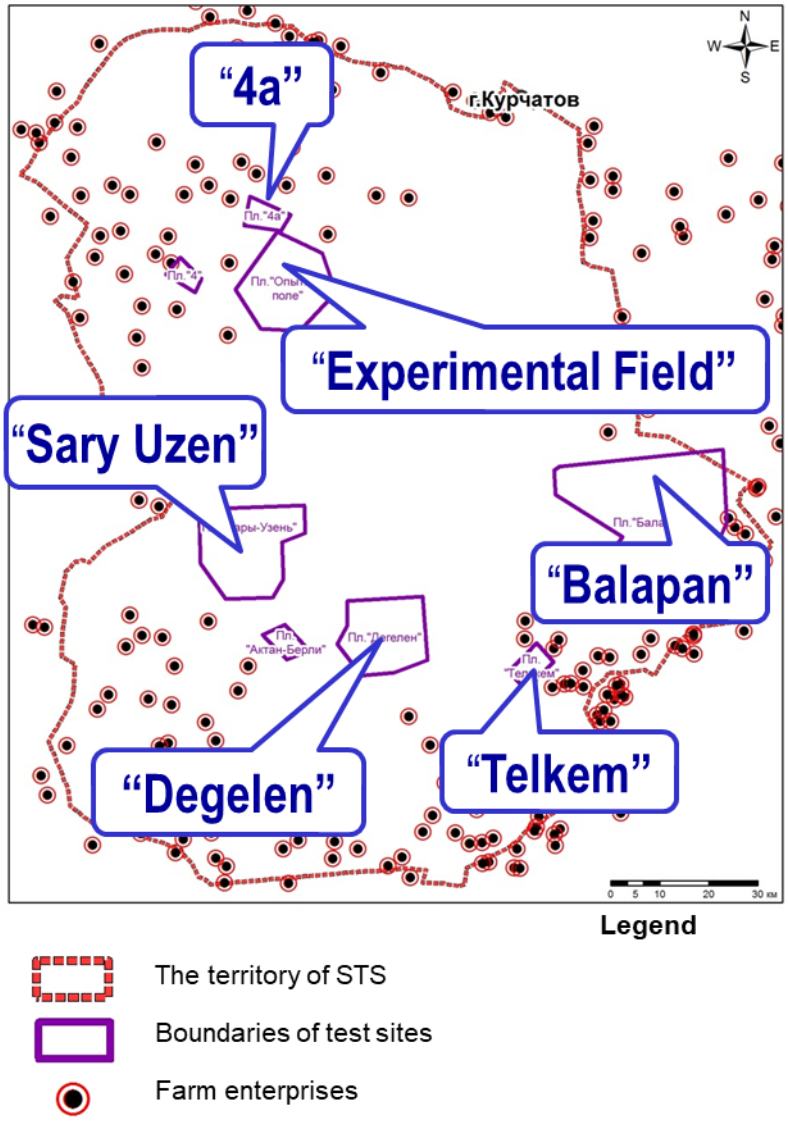


Figure 6.

Schematic Map of the STS and the location of farm enterprises and testing areas

As shown in Table 14, the concentrations of radionuclides 137Cs and 90Sr in the meat and milk samples were below the detection limit using the equipment and methodologies employed. It may seem surprising that products obtained from the STS contain such low concentrations of radionuclides. However, it is essential to understand that the contamination in the area is localized or patchy, meaning that animals can graze in both "clean" and "polluted" areas throughout the day.

Table 14.

Activity concentrations of radionuclides in milk and meat samples

No.	Surveyed areas of the landfill	Products (n – number of samples)	Activity concentration of radionuclides, Bq kg-1		
			^{241}Am	^{137}Cs	^{90}Sr
1	district of the river Shagan	meat	–	–	–
		milk, (n= 2)	–	1.7±0.4	0.8±0.2
2	south-eastern part	meat, (n= 10)	<0.6	<0.8	–
		milk, (n= 12)	<0.4	<0.7	<0.09
3	south part	meat, (n=6)	<0.6	<0.8	–
		milk, (n=4)	<0.4	<0.7	–
4	southwestern part	meat, (n= 2)	<0.6	<0.8	–
		milk, (n= 3)	<0.4	<0.7	–

5.4 Assessment of the Contribution of Soil and Forage to the Daily Intake of Radionuclides into the Body of Animals at STS

The transfer coefficient is the principal parameter used to assess the level of radionuclide migration from the external environment into livestock products. To date, numerous scientific studies worldwide have explored the transfer of radionuclides to livestock products. Table 15 displays the values of the transfer coefficients for various agricultural products, as documented in the IAEA publication (1994, 2010).

Table 15.

Transfer Coefficients of Radionuclides into Livestock Products (IAEA, 2010)

Product Type	Transfer Coefficients (F_f) from feed per 1 kg (L) of product			
	Cs	*Sr*	*Am*	*Pu*
Horse meat	n.a.	n.a.	n.a.	n.a.
Beef	$\frac{3.0\times10^{-2}}{4.7\times10^{-3}-9.6\times10^{-2}}$ (58)	$\frac{2.1\times10^{-3}}{2.0\times10^{-4}-9.2\times10^{-3}}$ (35)	$\frac{5.0\times10^{-4}}{-}$ (1)	$\frac{6.0\times10^{-5}}{8.8\times10^{-8}-3.0\times10^{-4}}$ (5)
Poultry Meat	$\frac{2.7\ *}{1.2-5.6}$ (13)	$\frac{2.0\times10^{-2}\ *}{7.0\times10^{-3}-4.1\times10^{-2}}$ (7)	n.a.	n.a.
Horse milk	n.a.	n.a.	n.a.	n.a.

Note – average values are provided in the numerator, the range of values in the denominator, and the number of F_f values in the IAEA database is indicated in parentheses;
"n.a." indicates data not available. * - Includes values for duck

The data indicate that the transfer of *Cs* and *Sr* radionuclides into livestock products has been extensively studied. Studies on the transfer of Pu and Am into livestock products are extremely rare. However, applying this data may be inappropriate under our conditions because the transfer coefficients vary widely. Additionally, most studies were conducted in laboratory settings or after radiation incidents in the Southern Urals and the Chernobyl nuclear power plant. The conditions at STS differ significantly in terms of radioactive contamination, animal breeds, productivity, feeding, housing conditions, forage plant types, and soil type, among other factors. It is also important to note that most research focused on the "feed–livestock products" pathway, whereas studies on the "soil/turf–livestock products" pathway were secondary. This is because feed is the primary source of

radionuclide entry into animals. However, given its steppe zonation, soil and turf can also contribute substantially to the radionuclide content in the daily animal diet.

Therefore, when assessing contamination levels in agricultural products, it is essential to use transfer parameters (e.g., transfer coefficients, half-lives) that were derived under the same conditions in which the products are generated.

It is important to note that no data were available for horse products, including horse meat and milk, at the time of the IAEA publication (2010). This gap underscores the significance of the current study, which contributes pioneering data on the transfer coefficients of Am and Pu in horsemeat, filling a critical void in the existing literature and offering new insights into the behavior of actinides in livestock products

Research Conducted at the STS

Field experiments were carried out to obtain transfer coefficients for animal-derived agricultural products within the STS agricultural complex, as detailed in Chapter 4 of this Thesis. The coefficients derived from the diet to livestock products under STS conditions are presented in Table 16.

Table 16.

Transfer Coefficients of Radionuclides into Livestock Products

Transfer Coefficients (F_f)	$^{137}Cs.$ $\times10^{-2}$	$^{90}Sr.$ $\times10^{-3}$	$^{241}Am.$ $\times10^{-5}$	$^{239+240}Pu.$ $\times10^{-7}$
Chicken meat				
Forage	192.9	–	–	–
Soil from ground test epicenters	18.8	–	7.5	–
Horse meat				
Forage (leachate-contaminated feed)	2.6	0.88	3.0	5.9
Soil from ground test epicenters.	0.013	0.24	0.68	1.4

The data from the table suggest that transfer coefficients vary by source. Radionuclides transfer more efficiently with water than with feed, and least efficiently with soil. Therefore, it is crucial to consider the contributions of each component separately when evaluating radionuclide transfer to livestock products.

Accepted Transfer Coefficients

The metabolism of radionuclides in animal bodies during field studies at the STS is comparable to existing data. The obtained transfer coefficients (F_f) for ^{137}Cs in chicken meat and horse meat were found to be nearly identical to those reported in the IAEA database (2010). The coefficients for $^{239+240}Pu$ under full-scale STS conditions were in the range of values in the IAEA technical document. However, worth noting the range of values presented in the IAEA data varies widely, spanning nearly four orders of magnitude. These discrepancies are due to a myriad of factors (e.g., natural and climatic conditions, species characteristics of the animals, radionuclide forms), and it is difficult to determine which factor is most influential.

For assessing radionuclide content in livestock products, the transfer coefficients determined in field studies at STS were utilized (Table 16). For products where current data are lacking, the average coefficients from the IAEA database were used. In cases where soil-to-product transfer coefficients were unavailable, coefficients from hay were substituted and vice versa. The Transfer Coefficient of Americium (Am) from feed to chicken meat was estimated to be two orders of magnitude higher than the Transfer Coefficient of this radionuclide from soil to chicken meat (the same as for Cs). In the case of Sr, the Transfer Coefficient from soil to chicken meat was estimated to be two orders of magnitude lower than the Transfer Coefficient from feed to chicken meat. Since the IAEA database did not provide data for horsemeat, data for beef were used instead. The coefficients used to calculate radionuclide content in livestock and poultry products are presented in Table 17.

5.4.1 Calculation of Expected Concentrations of $^{239+240}Pu$, ^{241}Am, ^{137}Cs, and ^{90}Sr in Products

Radionuclides can enter the bodies of animals through various pathways. However, it is understood that the ingestion, inhalation, or dermal absorption of radionuclides by animals grazing on contaminated land is likely to be negligible. The IAEA handbook provides essential data on the transfer parameters from forage

to livestock products such as milk, meat, and eggs. Nevertheless, it does not consider the transfer parameters from water or soil, which could significantly influence the level of contamination in the final product.

Table 17.

Accepted Transfer Coefficients for Calculating Radionuclide Content in Products

Product Type	Cs	*Sr*	*Am*	*Pu*
	F_f from feed per 1 kg (l) of product			
Horse meat / Beef	2.6×10^{-2}	2.1×10^{-3} *	3.0×10^{-5}	5.9×10^{-7}
Chicken meat	1,9	2.0×10^{-2}	*7.5×10^{-2}* **	-
	F_f from soil per 1 kg (l) of product			
Horse meat	1.0×10^{-4}	6.0×10^{-4}	$<2.0\times10^{-6}$	1.0×10^{-7}
Chicken meat	18.8×10^{-2}	*2.0×10^{-4}* **	7.5×10^{-5}	-

note – – data from field experiments at STS; XXX – IAEA data;
«–» – no data available. «*» – data for beef from IAEA.«**» – estimated data.

For a comprehensive prediction of radionuclide concentrations in livestock products, it is crucial to account for all potential contamination pathways. This includes not only the radionuclides present in forage but also those in soil and water. To accurately predict the concentration of radionuclides in livestock products (C_{prod}), we must consider the daily intake of radionuclides from all environmental sources and apply appropriate transfer coefficients (F_f) from the diet to the final products.

$$C_{prod} = V_{(soil)}\times C_{(soil)}\times F_{f\,(soil)} + V_{(forage)}\times C_{(forage)}\times F_{f\,(forage)} + V_{(water)}\times C_{(water)}\times F_{f\,(water)} \quad (5)$$

where:

V forage is the daily forage intake (kg day^{-1});

V soil is the amount of unintentionally ingested soil (kg day^{-1});

V water is the daily water consumption (l day^{-1});

C forage is the activity concentration of radionuclides in forage (Bq kg^{-1});

C soil is the activity concentration of radionuclides in soil (Bq kg^{-1});

C water is the activity concentration of radionuclides in water (Bq l^{-1});

F_f feed, F_f soil, F_f water are the coefficients of radionuclide transfer from forage, soil, and water to the products per kg or l of product, respectively.

In our calculations, the intake of radionuclides through water was not considered.

The migration of radionuclides through the soil–plant system is the initial stage of the biological cycle, crucial for determining the level of radionuclide transfer into animals and the products derived from them. The concentration of radionuclides (^{241}Am, ^{137}Cs, ^{90}Sr, *and* $^{239+240}Pu$) in plants can be derived using the experimental data from the study of radionuclide accumulation parameters by plants at the STS:

$$C_{(forage)} = C_{(soil)} \times T_{f\,(soil\text{-}plant)}; \qquad (6)$$

where $T_{f\ (soil\text{-}plant)}$ is the coefficient of radionuclide accumulation by plants, discussed in Chapter 2.4.

The daily intakes of feed and soil are based on literature data (Table 18) (Kalashnikov et al. 1985). The calculation of radionuclide intake from soil ingested during grazing was based on the observation that cattle can consume up to 600 kg of soil, and sheep up to 75 kg of soil during the pasture period (Korneev, N.A., Sirotkin, A.N. 1987; Alexakhin, R.M., et al. 1992). Broilers reared in free-range conditions, whether on grass-covered plots or under trees, were found to ingest small amounts of soil daily. The amount of soil ingested varied, but generally did not exceed 3 grams per day. However, in adverse conditions such as winter, older birds under trees could ingest up to nearly 5 grams of soil daily (Jurjanz et al., 2015).

Table 18.

Physiological Parameters of Farm Animals Used for Calculation

Indicator	Horse / Cattle	Broiler
Daily forage intake, kg day^{-1}	15	0.011
Amount of soil inadvertently ingested, kg day^{-1}	1.6	0.003

In our calculations, we analyzed two distinct areas within the STS: the conditionally 'background' territories and the Ground Test Site ('Experimental Field'). The descriptions of these areas are provided in Chapter 2.1. In each area, we selected the highest activity concentrations of radionuclides present in the soil. The activity concentration of radionuclides was calculated based on the transfer coefficient in the soil-plant system for both the 'background' territories and the Ground Test Site. The accumulation coefficients of radionuclides, including ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am, by plants at different STS sites have been presented by Larionova et al. (2018, 2021). Average Activity concentration of Radionuclides in Soil and Plants presented in Table 19.

Table 19.

Average Activity concentration of Radionuclides in Soil and Plants

Source	$^{239+240}Pu$, Bq kg^{-1}	^{241}Am, Bq kg^{-1}	^{137}Cs, Bq kg^{-1}	^{90}Sr, Bq kg^{-1}
Conditionally "background" territories of the STS				
Soil (Lukashenko, S. N. 2013)	29,6	2,5	144,0	460,7
Forage	0,2	0,014	2,9	12,0
Ground test site ("Experimental Field")				
Soil (Data from tab. 3)	150 000	19 000	6000	6 700
Forage	210	10	17	154

The results of calculating the daily radionuclide intake into the bodies of farm animals during grazing on "background" territories and at the Experimental Field site are presented in Table 20.

Table 20.

Daily Intake of Radionuclides into the Body of Farm Animals in Conditionally Background Territories

Farm Animals	Live weight. kg	Radionuclide intake into the body of 1 animal. Bq							
		Daily intake from feed				**Daily intake from soil**			
		$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
Conditionally "background" territories of the STS									
Horse	350–400	3.0	0.21	43.5	180.0	47.4	4.0	230.4	737.1
Chicken	1.7-3.3	0.0022	0.00015	0.032	0.13	0.089	0.0075	0.43	1.4
Ground test site ("Experimental Field")									
Horse	350–400	3150	150	255	2310	240000	30400	9600	10720
Chicken	1.7-3.3	2.3	0.11	0.19	1.7	450	57	18	20

Based on this data, we can estimate the concentrations of radionuclides in livestock products produced in the study area. The expected concentrations are detailed in Table 21.

Table 21.

Expected Concentrations of Radionuclides in Livestock Products

Product Type	Predicted concentration of radionuclides in products from forage intake, Bq kg^{-1}				Predicted concentration of radionuclides in products from soil intake, Bq kg^{-1}			
	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
Conditionally "background" territories of the STS								
Horse meat	1.8×10^{-6}	6.3×10^{-6}	1.1	3.8×10^{-1}	4.7×10^{-6}	8.0×10^{-6}	2.3×10^{-2}	4.4×10^{-1}
Chicken meat	n.a.	1.2×10^{-5}	6.1×10^{-2}	2.6×10^{-3}	n.a.	5.6×10^{-7}	8.1×10^{-2}	2.8×10^{-4}
Ground test site ("Experimental Field")								
Horse meat	1.9×10^{-3}	4.5×10^{-3}	6.6	4.9	2.4×10^{-2}	6.1×10^{-2}	9.6×10^{-1}	6.4
Chicken meat	n.a.	8.3×10^{-3}	3.6×10^{-1}	3.4×10^{-2}	n.a.	4.3×10^{-3}	3.4	4.0×10^{-3}

The values of expected concentrations of radionuclides in agricultural products, when animals graze in conditionally "background" territories, are significantly lower than those shown in Chapter 5.2.

5.4.2 Calculation of Boundary Parameters $^{239+240}Pu$, ^{241}Am, ^{137}Cs, and ^{90}Sr in Soil

To determine where livestock can graze safely and products meet hygienic standards, we calculate the maximum permissible levels (MPL) of radionuclides ($^{239+240}Pu$, ^{241}Am, ^{137}Cs, and ^{90}Sr) in the soil. Based on formulas (5) and (6), it follows that:

Tables 22 show calculated MPLs of radionuclides in the soil for various sections of the STS, enabling the production of livestock products like horse meat and chicken meat that meet sanitary and hygienic standards.

$$MPL_{(soil)} = \frac{C_{PL\ prod}}{V_{(soil)} F_{f\,(soil)} + V_{(forage)} F_{f\,(forage)}\ T_{f\,(soil\text{-}forage)}} \qquad (7)$$

Table 22.

Maximum Permissible Levels of Radionuclides in Soil at Epicenters of ground tests of STS, MBq kg^{-1}

STS Sites	$^{239+240}Pu$	^{241}Am	^{137}Cs	^{90}Sr
Horse meat	116,0	5,8	0,16	0,29
Chicken meat	-	30,6	0,29	14,1

It has been demonstrated that grazing animals in areas with high concentrations of radionuclides in the soil, especially transuranic elements, can yield livestock products that meet the sanitary and hygienic standards for radiation (refer to Table 22). Therefore, the transfer of radionuclides into livestock products from conditionally 'background' territories is not considered a risk to economic activities.

With prolonged grazing in contaminated areas, the concentrations of radionuclides in the primary organs of deposition (^{90}Sr in bone tissue, ^{241}Am and $^{239+240}Pu$ in the liver) will be significantly higher than in the meat of these animals. However, the contaminated soil areas are relatively small compared to the entire sites. For instance, at the 4a test sites, which cover an area of 63 km², the soil contaminated with ^{90}Sr levels exceeding 1.5 kBq kg^{-1} spans only 8.0 km², and areas with contamination levels above 100 kBq kg^{-1} cover merely 0.14 km². The area of soil contamination with ^{241}Am and $^{239+240}Pu$ at the Experimental Field site (totaling 375 km²) that exceeds 10 kBq kg^{-1} does not surpass 1.5 km² (see Chapter 2.1). Therefore, the short-term presence of animals in these regions will result in negligible radionuclide contamination of livestock products, posing no significant risk when considering the dose of internal radiation to the population.

5.5 Assessment of the Contribution of Soil, Water, and Vegetation to the Daily Intake of Radionuclides into the Body of Animals at STS

When grazing animals within the territory of the agricultural complex, potential sources of radionuclides include air, water, soil, and vegetation. Previous studies (Baigazinov Zh.A. 2010) have determined that the contribution of air to the radionuclide intake in animals and birds is minimal and can be disregarded.

The primary vectors for radionuclide transmission into farm animals are soil, water, and vegetation. Soil entry into the animal body occurs through two main pathways: dust on vegetation and soil that animals inadvertently ingest from the ground.

The significance of soil and vegetation as sources of radionuclide intake varies considerably across different sites. Research conducted at the Experimental Field site indicates that over 85% of ^{90}Sr may originate from soil. The principal contributors of ^{137}Cs, ^{90}Sr, ^{241}Am, and $^{239+240}Pu$ into farm animals are soil and vegetation. The calculations detailing the proportion of each natural component in the daily radionuclide intake are presented in Table 23.

These variations are primarily due to the different forms of radionuclides present in the soil and the radionuclide transfer coefficients by plants, which are discussed in Chapter 2 (Sections 2.3 and 2.4).

Table 23.

Contribution of Soil and Forage to the Daily Intake of Radionuclides into the Body of Animals (e.g., horse, chicken)

on the Conditionally "background" territories of the STS, %

STS Sites	$^{239+240}Pu$		^{241}Am		^{137}Cs		^{90}Sr	
	forage	soil	forage	soil	forage	soil	forage	soil
Horse	6,0	94,0	5,0	95,0	15,8	84,2	19,6	80,4
Chicken	2,4	97,6	2,0	98,0	6,8	93,2	8,7	91,3

Note – Calculations were based on the radionuclide accumulation coefficients obtained from plants at the STS.

on the Epicenters of ground tests of the STS, %

STS Sites	$^{239+240}Pu$		^{241}Am		^{137}Cs		^{90}Sr	
	forage	soil	forage	soil	forage	soil	forage	soil
Horse	*1,3*	*98,7*	*0,5*	*99,5*	*2,6*	*97,4*	*17,7*	*82,3*
Chicken	*0,5*	*99,5*	*0,2*	*99,8*	*1,0*	*99,0*	*7,8*	*92,2*

Note – Calculations were based on the radionuclide accumulation coefficients obtained from plants at the STS.

Based on previously accepted physiological indicators of farm animals, calculations of radionuclide activity in soil and plants, and the daily intake of radionuclides into the bodies of farm animals, it is possible to estimate the contribution of soil and feed to the radionuclide contamination of livestock products. Depending on the nature and level of contamination at STS sites, the

contributions of these natural components to radionuclide entry into horse and chicken meat can vary significantly (Table 24).

Table 24.

Contribution of Soil and forage to the Contamination of Livestock Products at the Epicenters of ground tests of STS, %

STS Sites	$^{239+240}Pu$		^{241}Am		^{137}Cs		^{90}Sr	
	forage	soil	forage	soil	forage	soil	forage	soil
Horse meat	7,2	92,8	6,8	93,2	87,2	12,8	43,0	57,0
Chicken meat	-	-	65,6	34,4	9,4	90,6	89,4	10,6

Note – Calculations were based on the radionuclide accumulation coefficients obtained from plants at the STS.

The data in the table illustrate the differential contributions of soil and forage to the radionuclide contamination of horse and chicken meat at the STS. For horse meat, soil is the primary source of $^{239+240}$Pu and ^{241}Am, contributing 92.8% and 93.2%, respectively, compared to much lower contributions from forage (7.2% and 6.8%). Conversely, ^{137}Cs contamination in horse meat is predominantly from forage (87.2%), with soil contributing only 12.8%. ^{90}Sr shows a more balanced distribution, with 57.0% from soil and 43.0% from forage.

In chicken meat, ^{241}Am and ^{137}Cs are primarily derived from forage, with contributions of 65.6% and 9.4%, respectively, while soil accounts for 34.4% and 90.6%. For ^{90}Sr, forage again dominates with 89.4%, compared to 10.6% from soil. These results highlight that the source of radionuclide contamination varies significantly depending on the specific radionuclide and the type of livestock, emphasizing the need to consider both soil and forage in contamination assessments.

5.5 Conclusion on Chapter 5

The research findings suggest that grazing animals directly on the STS test sites, where soil and vegetation contamination levels can be substantial, will result in livestock products that comply with the hygienic standards of the RK (Order No-71, 2022). This compliance is primarily due to two factors: first, the contamination areas at these test sites are small (only 1.0-1.5 km²) and localized, despite the fact that horses cover approximately 15-30 km of pasture territory per

day. Second, the migration capacity of radionuclides from soil to feed to products is relatively low in the soil of the STS territory.

In general, it is expected that any type of livestock product obtained from the agricultural complex outside the test sites (i.e., over 95% of the territory of the STS) will meet Hygienic Standards of the RK (Order No-71, 2022).

With further studies aimed at reintegrating the STS territory into economic circulation, the assessment of livestock product quality may primarily rely on theoretical calculations. Despite feed being the typical principal source of contamination, under specific conditions (such as various types of testing, high localization of pollution, and unique soil and climatic features), the main pathway for radionuclide entry into the bodies of productive animals is through the direct ingestion of soil.

SUMMARY

This PhD thesis investigates the behavior of radionuclides, specifically ^{241}Am and ^{137}Cs, in livestock, focusing on horses and broilers exposed to contamination from the STS in Kazakhstan. The research encompasses three key studies: the transfer of radionuclides to horse tissues, the transfer to broiler tissues, and the subsequent excretion of these radionuclides from broiler tissues after long-term exposure. The findings show that radionuclide transfer is significantly influenced by the source of contamination, with higher transfer rates observed from contaminated feed compared to soil ingestion. The study also reveals that ^{137}Cs is rapidly excreted from animal tissues, following an exponential decay pattern, whereas ^{241}Am is retained much longer, particularly in liver and bone tissue. These results provide crucial data for assessing the potential risks associated with consuming livestock products from contaminated regions and underscore the importance of ongoing monitoring and research in nuclear-affected areas.

Thesis 1.:Radionuclide Transfer and Retention: My studies highlight that the transfer of ^{241}Am, ^{137}Cs, ^{90}Sr, and $^{239+240}$Pu to animal tissues varies significantly depending on the source of contamination (soil vs. contaminated feed). For horses, the highest transfer coefficients (F_f) were observed in the liver of mares fed leachate-contaminated feed, with values reaching (72±22) $\times10^5$ d kg^{-1} FW for ^{241}Am and (31.8±8) $\times10^5$ d kg^{-1} FW for $^{239+240}$Pu. In fillies, the highest value for ^{137}Cs was 35.3×10^{-3} d kg^{-1} FW, while for ^{90}Sr, the highest was recorded in the ribs, reaching $(720\pm144)\times10^{-3}$ d kg^{-1} FW. These findings include pioneering data for ^{241}Am and $^{239+240}$Pu transfer coefficients in horsemeat at the STS, marking the first such reports both locally and globally. The IAEA handbook (2010) does not provide transfer coefficient values for horses, making this study a significant contribution to the understanding of actinide transfer in livestock.

Thesis 2.:Transfer Parameters: The transfer parameters of radionuclides ^{137}Cs and ^{90}Sr into the organs and tissues of yearling fillies were found to be higher than in adults by up to 1.6 and 3.6 times, respectively. However, I have found that age does not appear to significantly affect the accumulation of $^{239+240}$Pu and ^{241}Am in

horse tissues. In broilers, the Transfer Coefficient for ^{137}Cs in muscle from the grass meal diet was significantly higher at (192.9±25.8) ×10^-2 d kg^{-1} FW, compared to (18.8±4.6) ×10^{-2} d kg^{-1} FW from soil contamination. For ^{241}Am, the Transfer Coefficient in muscle was (1.1±0.95) ×10^{-4} d kg^{-1} FW.

Thesis 3.:Dynamics of Accumulation: I have identified for the first time the accumulation dynamics of ^{137}Cs and ^{241}Am in boilers using contaminated feed from the STS territory. The study observed dynamic equilibrium stages for ^{137}Cs in broilers' muscle by the 30th day and in their liver and bone by the 14th day of feeding. For ^{241}Am, the muscle reached equilibrium immediately, while the liver and bone took longer to stabilize, with significant accumulation over the study period.

Thesis 4.:Excretion Dynamics: I have identified for the first time the excretion dynamics of ^{137}Cs and ^{241}Am in boilers using contaminated feed from the STS territory. The excretion of ^{137}Cs from broiler muscle, liver, and bone followed an exponential decay, characterized by an initial rapid phase followed by a slower phase. The majority of ^{137}Cs was excreted within the first week. Specific equations were used to model the excretion kinetics for each organ. By the 70th day, 20-50% of the initial ^{241}Am concentration remained in the liver, while approximately 35% persisted in the bone, demonstrating a more stable retention in bone compared to a gradual decline in the liver.

Thesis 5.:Soil as a Contamination Source: In areas with ground nuclear test epicenters, such as the STS, I have found that soil directly serves as a significant source of radionuclide contamination in livestock products, which pathway should be incorporated into international best practices. Updated data indicate that for horse meat, soil is the dominant source of $^{239+240}$Pu and ^{241}Am, contributing 92.8% and 93.2%, respectively, while the contribution from forage is substantially lower at 7.2% and 6.8%. Conversely, ^{137}Cs contamination in horse meat is primarily from forage (87.2%), with soil contributing only 12.8%. For ^{90}Sr, the distribution is more balanced, with 57.0% from soil and 43.0% from forage.

Thesis 6.:Food Safety Implications: The findings provide critical data for estimating internal radiation doses from the consumption of livestock products in regions impacted by nuclear tests. I have identified that the high retention of ^{241}Am in bone tissue and liver, in particular, poses a potential long-term risk to consumers, emphasizing the importance of monitoring and mitigating contamination in nuclear-affected regions.

Recommendations for Future Work

To build upon the findings of this research, several recommendations for future work are proposed:

1. Long-Term Monitoring: Establish a long-term monitoring program to continuously assess radionuclide levels in livestock products from the STS and other contaminated regions. This will provide ongoing data to refine risk assessment models and ensure the continued safety of food products.
2. Broader Species Analysis: Expand the study to include additional livestock species and a wider range of environmental conditions. This will enhance the robustness of the risk assessment models and provide a more comprehensive understanding of radionuclide transfer dynamics across different animal species and environmental contexts.
3. Advanced Mitigation Strategies: Develop and test advanced strategies for mitigating radionuclide transfer to livestock products. This could include innovative agricultural practices, soil remediation techniques, and feed additives designed to reduce radionuclide uptake by animals.
4. Detailed Mechanistic Studies: Conduct detailed mechanistic studies to better understand the biological processes governing radionuclide uptake, distribution, and excretion in livestock. Such studies could reveal new insights into how radionuclides interact with animal physiology and inform the development of targeted interventions.
5. Regulatory Framework Development: Collaborate with regulatory bodies to develop and implement evidence-based standards for permissible

radionuclide levels in livestock products. This will involve translating the scientific findings into practical guidelines and policies that can be applied in contaminated regions worldwide.

6. Public Health Education: Engage in public health education initiatives to raise awareness about the risks of radionuclide contamination in livestock products and promote best practices for minimizing exposure. This could include community outreach programs, informational campaigns, and the development of educational materials for farmers and consumers.

By addressing these recommendations, future research can build upon the foundations laid by this dissertation, further advancing the field of radioecology and contributing to the safe and sustainable management of agricultural practices in regions affected by nuclear contamination.

LIST OF PAPERS INCLUDED

This thesis is based on the following papers, which are referred into the text by the name of authors:

1. **Baigazinov ZhA**, Lukashenko SN, Panitsky AV, et al. **(2020)**. The transfer of $^{239+240}$Pu, ^{241}Am, ^{137}Cs and ^{90}Sr to the tissues of horses. *J of Env Radioactivity* (WoS Q3, Impact Factor 2.161) doi.org/10.1016/j.jenvrad.2020.106322

2. Mamyrbayeva AS, **Baigazinov ZhA**, Lukashenko SN, et al. **(2021)** The excretion of ^{241}Am and ^{137}Cs from the broilers organs after long-term application. *J of Env Radioactivity* (WoS Q3, Impact Factor 2.161) doi:10.1016/j.jenvrad.2021.106543

3. Mamyrbayeva AS, **Baigazinov ZhA**, Lukashenko SN, et al. **(2020).** The transfer of ^{241}Am and ^{137}Cs to the tissues of broilers' organs. PLoS ONE. (WoS Q1, Impact Factor 2.740) doi.org/10.1371/journal.pone.0235109

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