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Determination of Cs-137 Concentration in Some Environmental Samples around the Semipalatinsk Nuclear Test Site in the Republic of Kazakhstan

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Authors' contributions

This work was carried out in collaboration between all authors. Authors SD, Nadir Ibragimov and EO designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors AS, YR and OG managed the analyses of the study. Authors NB, Nurgul Ikimbayeva and MB managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Cs-137 was measured in the soil, vegetation, water, milk and meat samples taken from five different settlements around the Semipalatinsk nuclear test site (SNTS) (Kazakhstan). The settlements of Sarzhal, Kainar, Akzhar, Novopokrovka and Karkaraly belong to extreme, very high, very high, high and minimal radiation risk zones, respectively. Radionuclide activities concentrations were determined using a pure Ge gamma-spectrometer (Canberra, USA) and the measured activity levels were not found to exceed the maximum allowable limits 10 Bq/kg for water and 1000 Bq/kg for meat and milk according to the guidelines of World Health Organization. The highest activity concentration levels of Cs-137 were found in soil while the lowest levels were measured in water. Vegetation, milk and meat showed intermediate levels. In soil, the highest

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values were obtained on samples collected in Sarzhal (35.0±1.0 Bq/kg) and Kainar (23.0±1.0 Bq/kg). Sarzhal also showed the most elevated levels in vegetation (2.2±0.1 Bq/kg), milk (1.8 ± 0.6 Bq/kg) and water (0.05 Bq/kg). Kainar showed the most elevated level in meat (1.8±0.6 Bq/kg).

Keywords: Cs-137; soil; vegetation; water; milk; meat; East Kazakhstan; Soviet Union.

1. INTRODUCTION

The Semipalatinsk nuclear test site (SNTS) has been contaminated by radionuclides mainly as a result of atmospheric, aboveground and underground nuclear tests that were carried out over 40 years from 1949 to 1989. The Soviet Union conducted 456 nuclear explosions at SNTS, including 116 aboveground and 340 underground explosions [1]. This led to radioactive contamination spreading not only inside the test site but also outside, where people live.

Survey of residual radioactivity in the soil at ten SNTS areas showed that a great number of the long-lived radionuclides Pu-239, Pu-240, Sr-90, and Cs-137 are concentrated in the soil at a depth of 0-8 cm [2]. Some of the residual radioactivity in the SNTS is tightly bound to the soil particles because of the extreme heat and resulting melting of the soils during the tests. The largest amount of radionuclides seems to have accumulated on micron-sized soil particles [3,4]. Wind erosion is responsible for the suspension of contaminated soil particles in the air and for spreading the contaminants [5,6]. For instance, dust particles of diameter 0.05-0.10 mm were reported to settle within a couple of kilometers of the erosion site, while particles of about 0.005-0.010 mm diameter were reported to travel 100 - 1000 km. The soil phase consists of mineral particles of various sizes, shapes and organic matter content in various stages of degradation [7,8]. The Cs-137 activity concentrations at Dolon and the adjoining three villages of Mostik, Cheremushka and Budene, near the SNTS territory, were found to be within 140.0 and 10.31 Bq/m^3 at 30 and 100 cm depths [9].

The radioactive contamination of soil can be due to anthropogenic (Sr-90 and Cs-137) and naturally occurring (U-238 and Th-232) radionuclides. It enters the human food chain via agricultural products [10,11]. An investigation conducted in 2001 has suggested that the most contaminated sites in the southeastern part of the Semipalatinsk test site are near the underground nuclear explosion sites Telkem-1 and Telkem-2 [12]. Soils in a radius of 600 m from the epicenter of the explosions were contaminated with the fission products Cs-137 and Sr-90, the activation products Am-241, Co-60, Eu-154, Eu-152 and the components of nuclear weapons material Pu-239/240 [12].

In these areas, vegetation that grows on the fields and meadows is the main source of radionuclides contamination transfer to animals [13] and, therefore of course, meat and milk. A cattle can consume the vegetation over a large area (up to 160 m^2) and about 1% of the Cs and the Sr consumed during the day are removed from the body through the milk [13]. Contamination of water and food products derived from farmed animals has the potential to represent a major source of radiation exposure to humans [14,15,16].

The aim of this study was to describe the distribution of Cs-137 contamination in soil, water, vegetation, milk and meat and to discuss the implications of such levels of contamination.

2. MATERIALS AND METHODS

2.1 Study Area

All sampling locations including Sarzhal, Kainar, Akzhar, Novopokrovka and Karkaraly, received inputs from the radioactive fallout due to the nuclear explosions conducted on SNTS. These inputs were largely from atmospheric and aboveground nuclear tests carried out on the “Experimental Field” test site and underground testing conducted elsewhere (Degelen, Balapan and Sary Uzen) [17]. The Balapan area (780 km^2, site of 105 undeground nuclear tests) may be the receptor of much of the SNTS radionuclide contamination, including the “Experimental Field” where 86 nuclear tests were conducted in the air and 30 on the ground [18]. This is because the dominant wind direction is from the West to the East and the underground water tends to run towards the East from the “Experimental field” region towards the Sarzhal and Kainar settlements [19]. The testing carried out at the “Experimental Field” site also contributed to the contamination of adjacent territories, including the Akzhar, Karkaraly and Novopokrovka settlements.

Samples of soil, water, vegetation, milk and meat were collected at 5 locations: Sarzhal, Kainar, Akzhar, Novopokrovka and Karkaraly. A description of the sampling locations is provided below. In accordance with the law of the Republic of Kazakhstan (from the 18th of December, 1992) "About social protection of citizens suffering from the consequences of nuclear testing in the Semipalatinsk Nuclear Test Site" adjacent territories belong to different zones of radiation risks.

1. The Sarzhal settlement, is situated 150 km South-West of the city of Semey and 25 km from the South-east border of the SNTS. This settlement belongs to the zone of extreme radiation risk. For this area, the most important contamination event was the testing of the first thermonuclear explosion that took place on the 23rd of August, 1953 [18]. Other potential sources of contamination, for the population of this village, include the explosion sites of Telkem-1 and Telkem-2 (where more than 200 underground nuclear explosions were carried out in the tunnels) situated several dozen of kilometers from Sarzhal [20,21].
2. The Kainar settlement, which is situated 275 km South-West of the city of Semey and 70 km South of SNTS, belongs to the zone of very high radiation risk. The main sources of radioactive contamination of the Kainar settlement are from the powerful aboveground nuclear test conducted on the 12th of August, 1953, the local aboveground nuclear test conducted on the 24th of September, 1951, the underground nuclear tests conducted on the 5th of October, 1954 and on the 16th of March, 1956 as well as the Telkem nuclear tests [18].
3. The Akzhar settlement (Pavlodar region) is situated 250 km North-West of the city of Semey and 80 km North of the SNTS. It belongs to the zone of very high radiation risk. The main potential source of contamination is from the nuclear tests which were carried out on "The Experimental Field" site. The Experimental Field was the first test site on SNTS and it was used for above ground and air nuclear tests between 1949 and 1962. The first nuclear explosion took place on the 29th of August, 1949. In total, 116 explosions tests were carried out. They included 86 in air and 30 above-ground explosions [1]. Test site area and adjacent territories were exposed repeatedly to radioactive contamination (the main sources were atmospheric explosions).
4. The Novopokrovka settlement (Borodulikha), which is situated 35 km South of the city of Semey and 165 km East of SNTS, belongs to the zone of high radiation risk. Again, the nuclear explosions conducted on the "Experimental field" were the main sources of contamination. This is because of the dominant wind coming from the West and blowing towards the East.
5. The Karkaraly settlement, which is situated 475 km West of the city of Semey city, 250 km North-East of the city of Karaghany and 120 km West of SNTS, belongs to the zone of minimal radiation risk.

All of these settlements are shown in Fig. 1. The number of livestocks kept by the settlements is shown in Table 1.

Table 1. Number of livestock kept by the settlements (2012)

Settlement	Cow	Sheep	Horse
Sarzhal	6726	36627	5648
Kainar	3500	11000	1800
Novopokrovka	2156	4062	186
Akzhar	1205	2540	150
Karkaraly	2300	7200	1200

2.2 Sampling and Analysis

According to the schedule plan, soil, water and vegetation samples were collected for radiochemical analysis. The collection took place in March and April 2012.

Vegetation was collected over an area that was not less than 6 m^2 (2 m x 3 m). Scissors sickle or knife were used to collect the top part of the vegetation leaving the bottom part of the plants (at least 3 cm from the ground). The vegetation cover on the studied areas is represented by feather grass (*Stipa capillata, S. sareptana, S. lessingiana)* and fescue (*Fectuca valesiaca*). After collection, the vegetation was stored at room temperature in draught cupboard. Samples were milled to pieces 1 cm long pieces and dried at 70 to 80℃ for 15 to 18 hours.

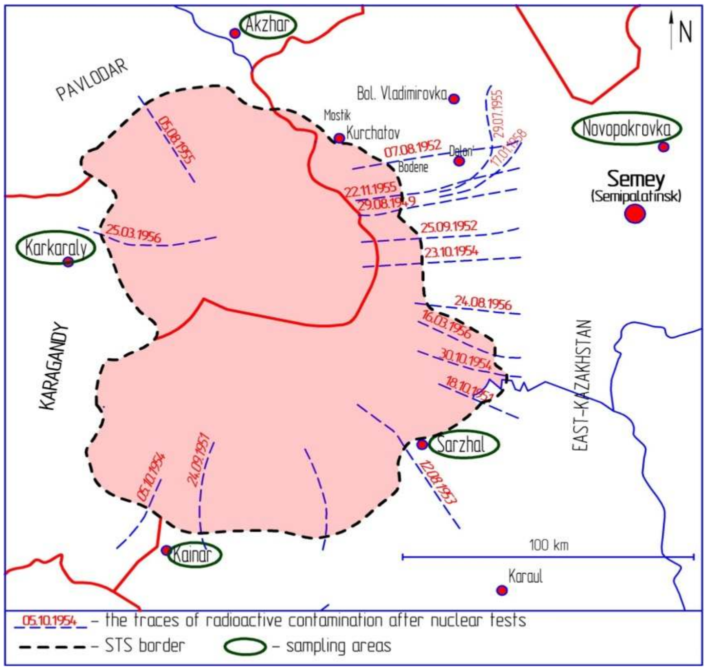

Fig. 1. Sampling locations (circled) with respect to the SNTS
(Latitude 50.30°, longitude 77.44°)

Soil was collected using a special shovel to a depth of 5 cm, because of the Cs-137 was expected to be found within the top (3–5 cm) of soils. For each grabs, the area sampled was 100 cm^2. To make up one sample, soil grabbed from 5 different places, within a 50 m perimeter, were mixed together in a large container. The soil samples were packed into polyethylene sack, labeled and transported to the laboratory. Four samples of 2 kg weight (totally 20 samples of 80 kg of soil) from each settlement were collected for radiochemical analysis.

For determination of the radionuclides in the soil and vegetation samples, the samples were left to dry for two weeks at room temperature. During that time, the soil was scattered on a rigid paper and mixed periodically. Next, the soil samples were milled and sifted through a 2 mm mesh sieve. The vegetation samples were milled using a blender. Then, the samples were transferred to a Marinelli vessel measuring container. For each analysis, the weight of the sample varied as the sample was prepared by volume (volume necessary to fill the Marinelli vessel). The vessel was then placed in the gamma-spectrometer.

Water samples were collected from the surface of the stream with a polyethylene or a metallic bucket. Care was taken to collect clear water and avoid collecting water containing suspended materials (including sediments). Water samples were filtered (filter pore size of 8 µm) and preserved with nitric acid (3 ml per 1000 ml of sample (pH<1)). The pH was verified using an indicator paper. Samples were stored for 5 days at a temperature of 2 to 5℃, in the dark room. Then, for each sample the radionuclide content was determined by the gamma-spectrometer.

From each settlement, in March-April 2012, samples of meat were obtained from 3 different cows (1 kg of meat from each cow). Also, milk samples (1 to 1.5 l) were collected from 3 cows. In order to avoid spoilage, meat samples were wrapped with gauze, moisten with 4-5% of formalin solution and packed into a polyethylene sack. Meat and milk samples were stored in the freezer at -18° to -200° C. Meat and meat products samples were cut into small pieces before being milled using an electric meat grinder. They were then placed into the Marinelli vessel and their radionuclide contents were determined by gamma-spectrometer. Collected milk samples were poured into clean glass airtight containers. Milk samples were preserved by addition of a 40% formalin solution (1-2 ml/l). Milk samples were poured into the Marinelli vessel and their radionuclides content was determined by gamma-spectrometry.

The Gamma-spectrometer used for Cs-137 determination was a pure Ge detector GC-2019 (55 cm^3) Canberra Company (USA). The Marinelli vessel characteristic were as follows: volume 1l, diameter 15 cm, height 110 cm. The calibration of the detector was done using a standard solution of Cs-137, Am-241 and Eu-152 dissolved in water. The activity of the solution was 10 Bq/l.

Surface radiation exposure dose rate were evaluated by dosimeter-radiometer RKS-01-SOLO (Solo LTD, Kazakhstan) with next technical characteristic: alpha flux density measurement range from 0.2 to $1{\cdot}10^5$ freq/min*sm^2, beta flux density measurement range – 2.0 to $1{\cdot}10^5$ freq/min*sm^2 gamma radiation intensity measurement range from $1{\cdot}10^2$ µSv/h to $1{\cdot}10^6$ µSv/h.

3. RESULTS AND DISCUSSION

The places with high exposure dose rate are identified as sampling points for soil and vegetation for further analysis. The average radiation exposure dose rate in the zone of extreme radiation risk, including Sarzhal settlement with a value of 0.32 µSv/h, while in the zone of high radiation risk the average dose rate were 0.2 µSv/h in Kainar, 0.13 µSv/h in Akzhar and 0.08 µSv/h in Novopokrovka settlements respectively. The flux density of alpha particles varied from 0.2 to 1.8 freq/min*sm^2. The flux density of alpha particles were 1.8, 1.45, and 0.32 freq/min*sm^2 in Sarzhal, Kainar and Akzhar respectively; while In Karkaraly, which belongs to the zone of minimal radiation risk, it was 0.2 freq/min*sm^2. The flux density of beta particles in all studied areas was under 10 freq/min*sm^2. Particularly, it was 8.7, 8.2 and 4.9 freq/min*sm^2 in Sarzhal, Kainar and Akzhar respectively. Table 2 and Fig. 2 present the results of the gamma analysis of tested samples.

The overall highest activity concentration (35.0 Bq/kg) was measured in the soil sample collected in Sarzhal, following by (23.0 Bq/kg)

Table 2. Cs-137 content in the samples

Sampling area	Cs-137 concentration (Bq/kg)				
	Soil	Water	Vegetation	Milk	Meat
Sarzhal	35.0±1.0	<0.05	2.2±0.1	1.8 ± 0.6	0.8±0.2
Kainar	23.0±1.0	<0.02	0.9±0.1	<0.6	1.8±0.6
Novopokrovka	1.8±0.2	<0.01	0.8±0.2	0.5±0.1	0.6±0.2
Akzhar	3.5±0.6	<0.03	1.3±0.2	<0.8	<0.2
Karkaraly	<0.5	<0.04	<0.3	<0.2	0.8±0.4

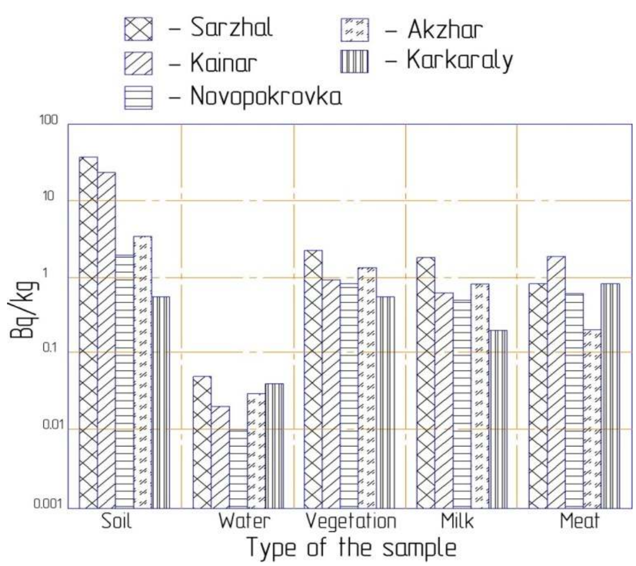

Fig. 2. Cs-137 activity concentration in soil, water, vegetation, milk and meat samples collected from 5 settlements surrounding the SNTS

obtained from the soil sample in Kainar. The other settlements showed Cs-137 activity concentrations that were much lower. Several studies were focused on the Cs-137 concentration in the area near to the SNTS. Priest et al. [7] reported, that the average concentration of Cs-137 in Sarzhal village (situated close to the south-east border of SNTS) was 27.0 Bq/kg. Sakaguchi et al. [9] studied the radiological situation in Dolon, Mostik, Cheremushka and Budene Settlements located north-east of SNTS and observed that Cs-137 content in soil varied from 140 Bq/m^2 to 10 310 Bq/m^2 (from 0.57 Bq/kg to 42.0 Bq/kg). Yamamoto and al. [4] analyzed the concentration of Cs-137 in soil sampled from the settlements, around the SNTS, which varied from 170 to 13 600 Bq/m^2 (0.69 Bq/kg to 55.77 Bq/kg).

Water analysis show that the activity concentration were not exceeding the maximum allowable concentration of 10 Bq/kg, set by WHO [22]. The measured levels were in the range of 0.01 to 0.05 Bq/kg with the highest level detected in Sarzhal. The results of the vegetation analysis showed the presence of low levels of Cs-137 in the samples. The highest values were obtained in Sarzhal (2.2 Bq/kq) and in Akzhar (1.3 Bq/kg), which were not exceeding the National maximum allowable limits for vegetation (74.0 Bq/kg) [23]. Again, in livestock products, Cs-137 activity concentration was not found to exceed the guidance level of 1000 Bq/kg reported in “General standard for contaminants and toxins in food and feed” (CODEX STAN 193-1995) set by Codex Alimentarius [24] and of 100 Bq/kg reported the Sanitary Rules of Republic of Kazakhstan [23]. However, milk samples were found to be most contaminated in comparison with other settlements in Sarzhal (1.8 Bq/kg) while meat samples were found to be most contaminated in Kainar (1.8 Bq/kg). The lowest values were obtained in Karkaraly’s soil, vegetation and milk samples. Akzhar was where the lowest meat value was obtained and Novopokrovka was where the water activity concentration was the lowest. Similar analysis of soil, vegetation and milk samples were performed by Kaimov et al. [14] reported that the highest levels of contamination were found in the Abai region (close to east border of SNTS), where the highest activity concentration of Cs-137 was 18.0 Bq/kg in soil, 7.60 Bq/kg in cow’s milk, 4.00 Bq/kg in the vegetation, and 3.00 Bq/kg in water. The lowest levels were measured within the Urdzhar region (classified in 1992 as being within the minimum radiation risk area), where 4.00 Bq/kg was found in the soil, 0.30 Bq/kg in the cow’s milk, 1.00 Bq/kg in the vegetation, and 0.20 Bq/kg in the water.

4. CONCLUSION

The concentration level of Cs-137 in samples of studied places decreases with the distance from the SNTS increases. The results revealed that the most contaminated settlements are Sarzhal and Kainar (classified in 1992 as being within an extreme radiation risk area). These settlements located near the south-east and south border of SNTS and were one of the first settlements which exposed radiation after conducting nuclear tests. A radioactive cloud resulting from these explosions moved with the wind to the south-east and south direction. The settlements, such as Novopokrovka, Akzhar and Karkaraly, located near the south and west border of SNTS are less contaminated. The contamination of the environment is negatively affecting to the sensory and biochemical characteristics of meat and meat products. All this can lead to lowering the biological and nutritional value of food and aftermath human diseases. Radionuclide transfer is a long-term and constant process. This work shows that the Cs-137 dose contribution from the consumption of livestock products is low in the 5 settlements and does not exceed the National limit and the limits approved by Codex Alimentarius. The data collected is part of a monitoring program that allows better estimates of radiation doses to population in some areas of concern. In the event that radiation risks would be deemed unacceptable, such data would also make it possible to identify mitigation options.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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