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Radionuclide contamination of underground water and soils near the epicentral zone of cratering explosion at the Semipalatinsk Test Site

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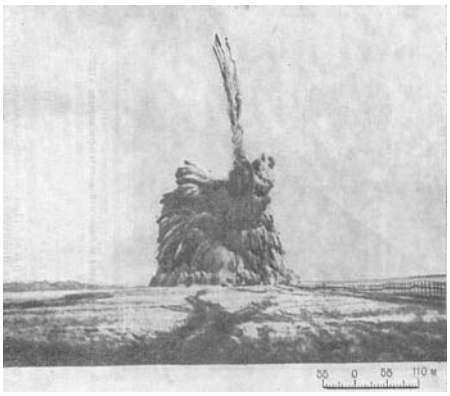
Abstract. The investigation wells for a control of the underground water contamination were bored after the cratering explosions at the Semipalatinsk Test Site, now they are restored partially. The analysis of the retrospective information of the Institute of Global Climate & Ecology (Moscow, Russia) give a possibility to choose wells and terrains for the successful study of radionuclide migration with the underground water. The epicentral zone, the crater and the territory with radius 1,5 km around the explosion "1003" were investigated under the ISTC project K-810. Underground water and soil samples were taken at the two expeditions of 2003. The chemical extraction methods taking into account the water mineral composition, gamma-spectrum methods, methods of liquid scintillation spectrometry and methods of alfa-spectrometry were used. The modern radionuclide content (^{3}H, ^{90}Sr, ^{137}Cs, $^{239+240}Pu$, ^{241}Am etc.) of the underground water is presented and compared with a radionuclide content of soils around crater. The retrospective information has been added by these modern data. The vertical radionuclide distrioin soils is presented.

1. INTRODUCTION

Researches of radionuclides migration with underground waters are especially important after producing of industrial underground nuclear explosions. Within the framework of project ISTC K-810 they were carried out at the Semipalatinsk Test Site on the example of underground cratering explosion "1003", 1965 (Fig. 1).

a) b)



a) 1965. Photo from [1]. b) 2003. Photo by E.Kvasnikova & S.Gordeev.

Figure 1. Underground cratering explosion "1003", Semipalatinsk Test Site.

A cratering explosion of 1.1 kt yield was conducted 48 m deep underground at the area named Sary-Uzen in October 14, 1965 (with reduced depth of 46.7 m/kt$^{1/3}$). Six seconds after the device was detonated the soil column reached its maximum height of 190 m, the column diameter being 240 m. It completely came apart 10-12 seconds after the explosion. The dust column rose as high as 300 m, mainly due to the atmospheric inversion layer [1].

According to the test plan, an extended charge of chemical explosives was detonated 2 hours after the nuclear explosion, in a trench radiating from the ground zero, to produce an experimental canal.

Parameters of irreversible deformation of the geological environment, based on data [1], are presented in Table 1.

Table 1. Zones of rock deformation around the "1003"explosion.

Zone of Irreversible Rock Mass Deformation	Radius, m
Folded zone	10
Crush zone	25
Intensive jointing zone	42.5
Block jointing zone (renewal of natural jointing)	70

Ground waters are confined within Paleozoic exogenous weathering rocks. Their depth of occurrence varies from 12 m to 20 m. Existing water-confining stratum - Neogene clays – makes the aquifer confined and creates water pressure in it.

The rock water-abundance ranges widely – from 0.4 m^3/day (Well 36) to 3.4 m^3/day (Well 22). The groundwater salinity varies within 1.7-3.3 g/l. As for the chemical composition, the following water types are found within the site: hydrocarbonate-sulfate magnesium-sodium, sulfate-chloride and chloride-sulfate sodium, sulfate sodium-magnesium and chloride-sulfate-hydrocarbonate sodium [3].

Ground waters basically discharged east and south towards the dry Saryuzyn-river bed. Its absolute heights range from 442 m to 441.6 m. Ground waters within the site are stagnant with the flow velocity of 0.13 m/day [2].

2. RETROSPECTIVE DATA ANALYSIS

Before explosion around the epicentral zone were bored wells for studying the subsequent migration of radioactive matters with underground waters from this zone. The first research has been carried out by the expedition leaded by Prof.Yu.Izrael & Dr.E.Stukin (Institute of Applied Geophysics under the State Hydrometeorological Sevice of the USSR) in 2 months and along one year after explosion [1]. Based on these retrospective data Fig.2 was compiled.

Four months after the explosion, the radioactive contamination moved north and south due to the asymmetry of the depression cone produced in the epicenter. The radioactivity of water samples taken from well 30 increased two orders of magnitude, and that of taken from well 20 one order of magnitude during 4 months of observation. East of the epicenter (wells 13, 15 and 45), the radionuclide content dropped down one-two orders of magnitude (Fig.2).

Five months after the explosion, the radioactive contamination within depression cone slightly increased from 6.8×10^{-12} Ci/l to 2.2×10^{-11} Ci/l (1.5-3 times in wells 28, 37 and 58). It was due to radionuclide desorption from induced jointing zones during rapid recovery of the piezometric surface. Because of the groundwater recharge in the northwest and south of the site, the total β-activity of samples collected from wells 22, 32, 33, 34, and 36 reduced from 7.9×10^{-11} Ci/l to 9.5×10^{-12} Ci/l.

The next observation period (6-12 months after explosion), the total β-activity gradually achieved equilibrium, and the radioactive contamination impoverished due to the general tendency towards the piezometric level recovery. A year later, the elevated total β-activities remained only east and south of the ground zero (wells 13 and 30).

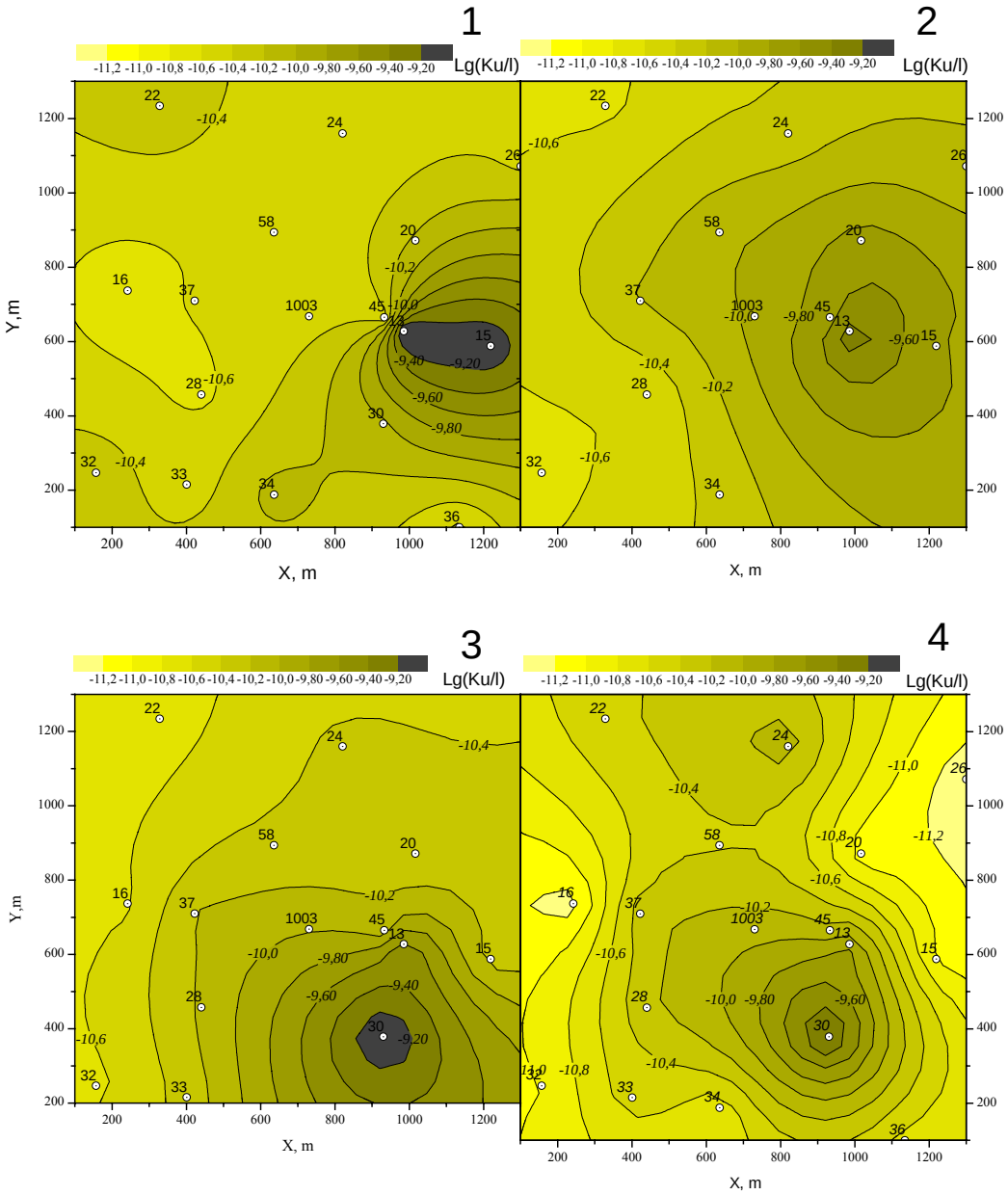


Figure 2. The change of the radiation situation around the place of "1003" explosion described based on the data received in 1965 by Prof.Yu.Izrael & Dr.E.Stukin (Institute of Applied Geophysics under the State Hydrometeorological Sevice of the USSR). Total β-activity, Ci/l.

3. FIELD AND LABORATORY INVESTIGATIONS

These wells have been cleaned and investigation of the radioactivity has been made after 35 years. From 15 observation wells, 13 were prepared for investigations (fig.3). Author's field investigation was carried out in March and October 2003. Underground water samples of (5-10) l have been taken by the cylindrical samplers.

When taking water samples from any horizon, the basic difficulty is to avoid well water contamination by soil surface radionuclides.

For this purpose the following rules has been used:

- sampling tool consisting of a sampling cylinder and a hoisting rope should be cleaned in the laboratory;
- sampling cylinder and hoisting rope should not touch the soil surface while taking a water sample;
- after every sampling the sampling cylinder should be placed onto a special clean rubber blanket or into a rubber bucket which prevents from contact with the soil surface;
- the hoisting rope should not touch the soil surface, too (a common tool known as winch meets this requirement);
- to control accuracy of water sampling, a soil sample should be taken within the well head area for radionuclide analysis and results of the soil sample analysis and analysis of the water solid residue after evaporation and suspension removal should be compared; presumably, the radionuclide ratio will show sampling quality and indicate necessity of re-sampling.

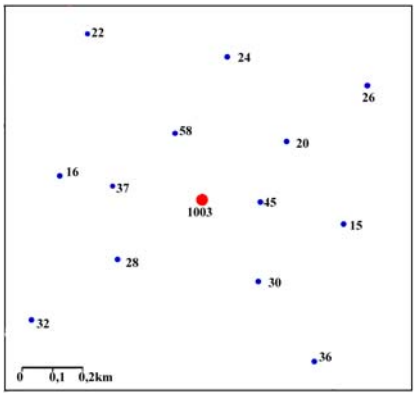


Figure 3. Geographical position of observation wells around the "1003" explosion.

Researches of soil near several wells have been carried out – to establish distinctions between radionuclide content of the underground waters contaminated at explosion, and of the soil contaminated due to the fallout from an atmosphere, generated the trace going from the epicenter to the North-East (45^0). Now this trace can be surveyed on 1.5-2 km from a place of explosion.

Soil samples were taken on a depth 0-5 cm near the wells (in 5-10 m). The soil section was described, the Brown semi-arid halomorphic soil was identified, and 6 soil layers (0-1, 1-2, 2-5, 5-10, 10-15 and 15-20 cm) were sampled in a fixed geometry. The description of soil is given in the table 2.

Table 2. Soil near the "1003"explosion.

Soil layer	Depth, cm	Description
A_d	0-1	Sod layer, free, grey, loamy, with roots, rubble
A_1	1-2	Humus layer, free, grey-brown, loamy, with roots, rubble
Bh	2-4	Illuvium layer with humus depot, color non-uniform, very dense, loamy, includes carbonates, rubble, with roots
$B_{Ca(Na)}$	4-12	Illuvium halomorphic layer, clear-brown, very dense, loamy, includes carbonates, rubble, with roots
B_{Ca}	>12	Illuvium layer, clear-brown, very dense, loamy, includes carbonates, rubble

The γ-spectrometry device type Canberra with a high pure Germanium detector with a 30% efficiency was used. To measure ^{241}Am X-ray spectrometer was used. Tritium is measured by beta spectrometry method using liquid scintillation detector.

Laboratory radio-chemical analysis of samples have been worked up:

- Dividing samples into suspended and liquid phases before laboratory analysis: a water sample taken from a well is added hydrochloric acid; the radioactivity in the sample is distributed between the liquid and suspended matter; a centrifuge is used to separate fractions.
- Radionuclide content analysis of samples, including a method for sample treatment before determination of ^{90}Sr and plutonium isotopes (^{239,240}Pu, ^{238}Pu, ^{241}Pu): radionuclides are concentrated by evaporation or co-precipitation of hardly soluble compounds, such as iron hydroxide, carbonate hydroxide or calcium oxalate; column chromatography is applied to separate and purify strontium; ion exchange and extraction were found to be mostly suitable for plutonium separation.
- ^{90}Sr and plutonium reduction in water samples, accounting of well's hydro-chemical characteristics: to enhance the technique sensitivity and take into account available results of salt composition analysis of liquid samples, a method of separate reduction of α-emitting radionuclides, including plutonium and strontium, from large-volume samples (up to 5 l) to various non-isotopic carriers has been proposed; iron hydroxide and calcium sulfate are used to reduce α-emitters and strontium, respectively.

4. RESULTS

Obtained results on the modern activity of the underground water are presented in the table 3. First of all the content of ^{3}H was detected, based on these data we could choose the informative wells. These are 6 from 13. In table 3 values on ^{3}H in samples from GGS-22, 37, 16, 32 are given for an example of non-informative situation. The ranges of values specified in the table 3 show distinctions in the radionuclide contents and fluctuations of their ratio in samples of water depending on a season that is connected with seasonal water's filling of wells.

Table 3. Contamination of ground water (sampling from wells), 2003.

Number of well	Co-ordinates from the epicenter	Radionuclide activity, Bq/l			
		^{3}H	^{137}Cs, x 10^{-3}	^{90}Sr, x 10^{-3}	$^{238+239}$Pu, x 10^{-3}
GGS-15	100^{0}, 510 m	202-324	105-2600	80	
GGS-24	10^{0}, 520 m	3	48-535	38-58	2.9-8.0
GGS-28	234^{0}, 370 m	50	131-2200	18-44	35
GGS-36	145^{0}, 730 m	102-255	222-1200	54-310	4.0-60
GGS-20	55^{0}, 360 m	>5-17	374-3100	20	
GGS-30	145^{0}, 370 m	1400-1700	469	104	40
GGS-22	325^{0}, 730 m	>5			
GGS-37	279^{0}, 320 m	>5			
GGS-16	279^{0}, 510 m	>5			
GGS-32	234^{0}, 740 m	>5			

Experimental and calculative relations of volatile and refractory radionuclides in soil samples and their depth distribution are given in the table 4. It is shown, that they are kept by constants up to depth of 10 cm. It allows to assume the presence of activity in the non-fractionated form, probably, as hot particles. On the experimental and calculative data deep distributions of activity of the investigated elements (fig.4-5) are compiled.

Receiving of non-fractionation's ratio from a soil hole enables to use them for the characteristic of underground waters contamination, and also to make the conclusion about receipt of a radio-activity in underground waters due to its migration of a cavity of explosion or due to secondary contamination from a ground surface.

Table 4. Radionuclide rations in soil samples, A_{152}/A_i, 2003.

Depth, cm	$^{152}Eu/^{137}Cs$	$^{152}Eu/^{60}Co$	$^{152}Eu/^{90}Sr$	$^{152}Eu/^{248}Pu$	$^{152}Eu/^{239+240}Pu$	$^{152}Eu/^{241}Am$
0-1	0.06	1.3	0.02*	1.5*	0.01*	0.06
1-2	0.05	1.1	0.02*	1.5*	0.01*	0.04
2-5	0.05	1.5	0.02*	1.5*	0.01*	0.06
5-10	0.05	1.4	0.02	1.5	0.01	0.05

*Based on the calculated values.

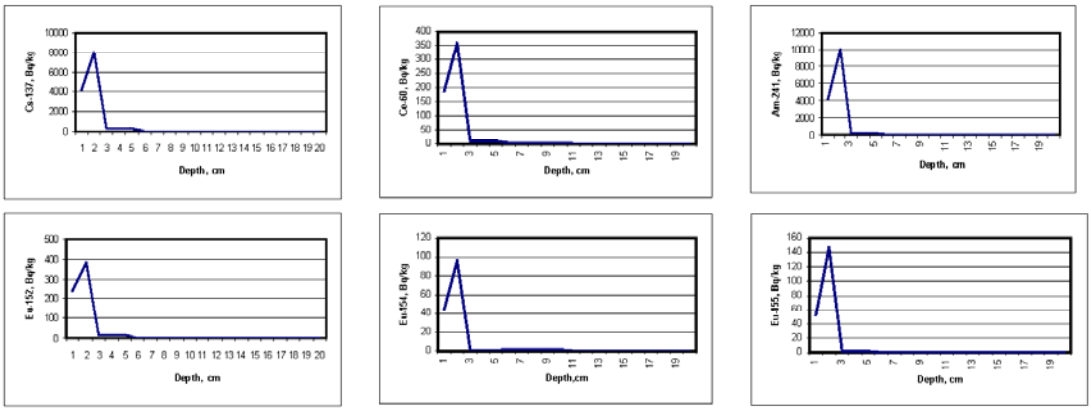


Figure 4. Depth distribution of ^{137}Cs, ^{60}Co, ^{241}Am, ^{152}Eu, ^{154}Eu, ^{155}Eu in the Brown semi-arid halomorphic soil near the "1003"explosion, 20 m from the edge of ground shaft around depression, 2003.

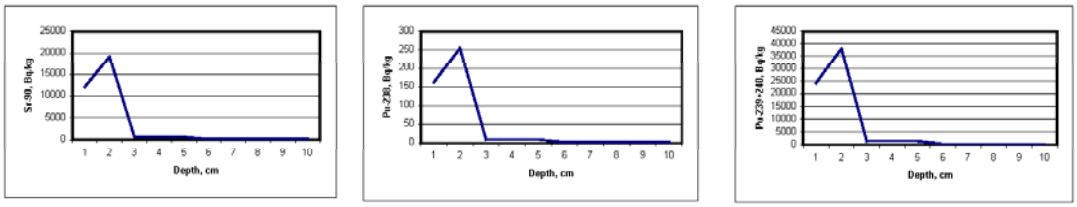


Figure 5. Depth distribution of ^{90}Sr and transuranium radionuclides in the Brown semi-arid halomorphic soil near the "1003"explosion, 20 m from the edge of ground shaft around depression, 2003.

5. CONCLUSIONS

1. Use of a complex method of the analysis of water and soil samples, including the steps described above, allows to determine radionuclide contamination for the answer on a question on migration of a radio-activity from a cavity of nuclear explosion in underground waters.
2. The analysis of radionuclide content of soil near the explosion's depression has shown preservation of ratio between volatile and refractory elements up to depth 10 cm. Such situation, probably, is caused by presence of radionuclides in the form of melting hot particles.
3. The analysis of ratio in water samples from wells shows significant fractionation aside enrichments by ^{137}Cs in comparison with a non-fractionation mix on a trace of atmospheric fallout.

References

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