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Radiological Situation in the Vicinity of Semipalatinsk Nuclear Test Site: Dolon, Mostik, Cheremushka and Budene Settlements

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Atomic explosion/Semipalatinsk/Dolon/Mostik/Cheremushka/Budene/^{137}Cs/^{239,240}Pu/Soil/Hot-particle.

The present situation of radioactive contamination at the village of Dolon and nearby villages such as Mostik, Cheremushka and Budene was investigated to serve as an aid to resolve dose discrepancy between model calculations and TL measurements made for external gamma-ray dose in air in Dolon. The paper was focused on the reevaluation of the accumulated levels and distribution of long-lived radionuclides ^{137}Cs and Pu isotopes in soil using long core samples up to a depth of 30 and 100 cm.

The inventories of ^{137}Cs and ^{239,240}Pu found were in the wide range of 140-10,310 and 140-14,320 Bq/m^2, respectively. Most of the Pu in soil was tightly incorporated into various sizes of fused particles. Both ^{137}Cs and ^{239,240}Pu in soil were accumulated in the smaller soil size fraction of <125 μm, and the presence of hot particles, probably due to Pu, was clearly observed by star-like patterns from α-tracks. The obtained data will be helpful for evaluating the current and future radiation risks to the people living around there.

INTRODUCTION

Dolon settlement is located about 100 km north-east of Semipalatinsk nuclear test site (SNTS), and is well known for receiving heavy exposure from radioactive fallout due to the first nuclear detonation on 29 August 1949. This explosion height was only about 40 m above the ground, and the energy output was equivalent to 22 kilotons of TNT. Incidentally, weather conditions of that day were unfavorable and storm-like. Two hours later, the radioactive cloud reached Dolon and nearby villages.[1,2] Therefore, centered on Dolon village, the radiological situation of external and internal doses, environmental contamination and body burden of humans has been studied by some investigators. According to Gusev, the population of this small village having an area of ca. 1.4 km^2 was about 1,300 at that time and agriculture was prosperous compared with today. Deriglazov *et al.*[3] reported that 90% of inhabitants received external effective doses up to 140 rem (1.4 Sv) during the first year. On the base of the data existing there now, there are no doubts about the relatively high radiation dose of the Dolon population, but some discrepancies in dose estimation among investigators for the village have been pointed out.[2,4,5] Therefore, further studies are needed.

As one of the available methods of retrospective dose reconstruction, there is a model calculation based on the long-lived radionuclide ^{137}Cs ($T_{1/2}$ = 30.17 y) in soil. Some investigators reported the contamination levels of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in the area of Dolon settlement.[6–11] But, the number of samples monitored was too scant to estimate the radiation dose around there. Furthermore, the assessment of plutonium isotopes (^{239,240}Pu and ^{238}Pu) is also important from the aspect of internal radiation. Such situation encouraged us to investigate in more detail the present situation of radioactive contamination by ^{137}Cs and Pu isotopes (^{239,240}Pu and ^{238}Pu).

In this paper, we report the current radiological situation at Dolon and its adjoining three villages Mostik, Cheremushka and Budene. Emphasis was placed on (1) reevaluation of accumulated levels and (2) distributions of long-lived radionuclides ^{137}Cs and Pu isotopes in soil using long core samples up to a depth of 30 and 100 cm, and (3) character-

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istics of Pu in soil by using sequential leaching, particle size discrimination and α-track methods.

MATERIALS AND METHODS

Soil samples

Soil cores up to a depth of 30 cm were taken at Dolon, Mostik, Cheremushka and Budene settlements during 2002–2003 (Fig. 1 and Table 1). Long cores of 100 cm in depth were also taken to get information on the widest conceivable accumulated levels of radionuclides under various situations (e.g., soil type, human activities, vegetation cover, *etc*). At each site, we confirmed latitude and longitude by the global positioning system (GPS). The 30 cm core samples were taken by inserting a stainless steel pipe (4.7 cm in diameter) into the ground. The samples were subdivided into five parts: 0–5, 5–10, 10–15, 15–20 and 20–30 cm. For the 100 cm core samples, a large hole (ca. 1 × 1 × 1 m) was dug in the ground. Along its side, two core samples were taken by using 10 cm long pipes from the surface to 60 cm in depth and 20 cm long pipes from 60 to ca.100 cm in depth. The eight segments were obtained as follows: 0–10, 10–20, 20–30, 30–40, 40–50, 50–60, 60–80 and 80 - ca.100 cm in depth. All samples were air-dried, and sieved through a 2-mm mesh screen to remove pebbles and grasses.

For one surface soil sample (about 50 g, 10 cm in depth) from Dolon village, particle size discrimination was performed simply by dispersing the sample in distilled water and passing the suspension through a set of standard sieves down to 32 μm in diameter. The suspension passed through the 32 μm screen was filtered by using a 0.45 μm pore size filter. The seven fractions obtained were as follows: <0.45, 0.45–32, 32–88, 88–125, 125–250, 250–500 and 500–2,000 μm.

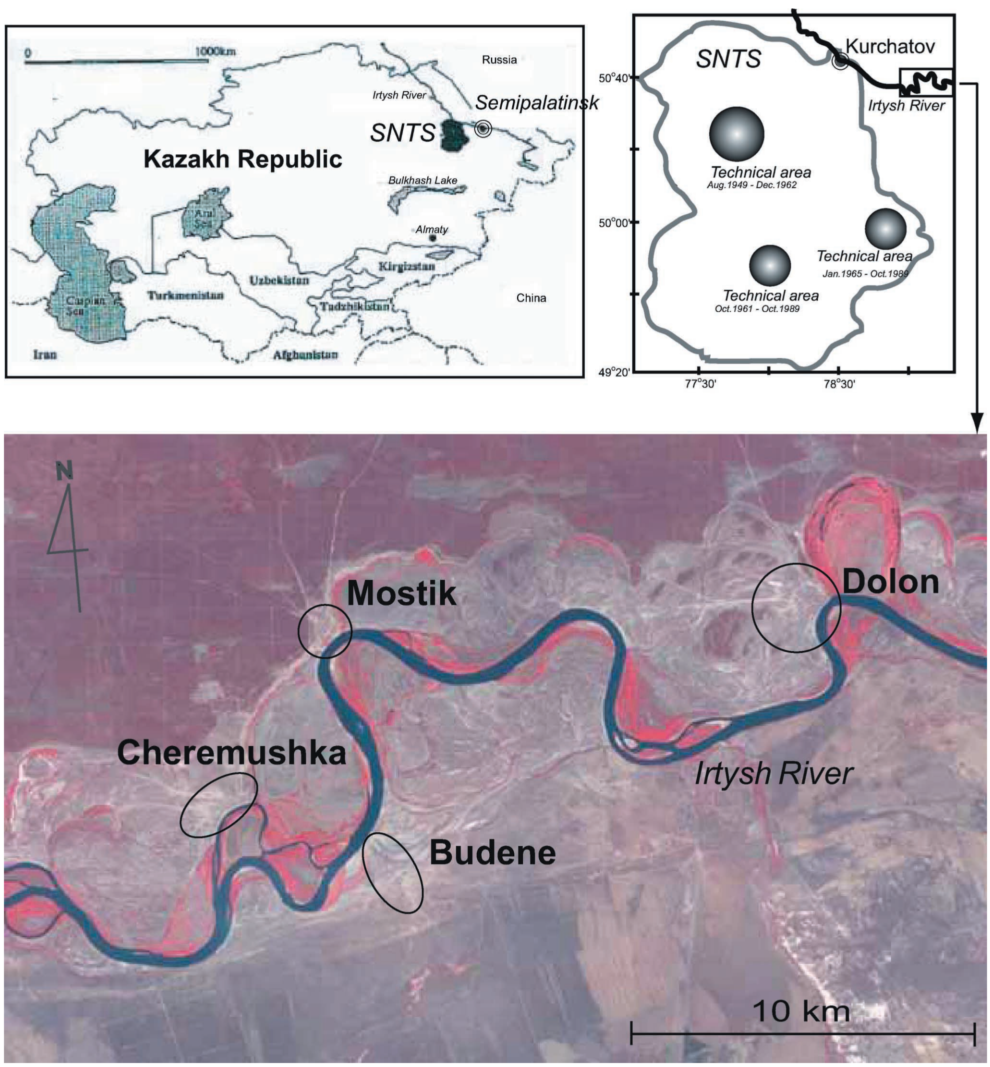

Fig. 1. Map around the Semipalatinsk nuclear test site, with the locations of Dolon, Mostik, Cheremushka and Budene settlements.

Table 1. Geographical position (by GPS) of sampling site.

Sampling site		Position		Remarks
Village	No.	N	E	
Dolon	D1	50°39’39”	79°18’42”	Small church in Dolon village.
	D2, 3	50°39’47”	79°18’56”	By the fence of cemetery at end of Dolon village. Appreciably sandy.
	D4	50°38’51”	79°18’14”	Seed stock house near forest (near Dolon).
	D5	50°39’42”	79°18’21”	By the fence of house in Dolon village.
	D6	50°39’42”	79°18’21”	By the fence of house in Dolon village.
	D7	50°39’38”	79°18’13”	By the fence of house in Dolon village.
	D8	50°39’38”	79°18’13”	By the fence of house in Dolon village.
	D9	50°39’45”	79°18’28”	By the fence of house in Dolon village.
	D10	50°39’42”	79°18’34”	By the fence of house in Dolon village.
	D11	50°39’53”	79°18’50”	By the fence of house in Dolon village.
	D12	50°39’38”	79°18’33”	By the fence of house in Dolon village.
	D13	50°39’49”	79°18’51”	By the wall of school in Dolon village.
	D14	50°41’ 0”	79°18’ 5”	Radioactive cloud center in plain (near Dolon). Hard ground.
	D15	50°41’ 0”	79°18’ 5”	Radioactive cloud center in plain (near Dolon).
	D16	50°40’35”	79°17’55”	Radioactive cloud center in plain (near Dolon).
	D17	50°40’33”	79°17’53”	Radioactive cloud center in plain (near Dolon).
	D18	50°40’ 1”	79°18’32”	By the fence of house in Dolon village.
	D19	50°40’ 0”	79°18’29”	By the fence of house in Dolon village.
	D20	50°40’ 3”	79°18’19”	By the fence of house in Dolon village.
	D21	50°39’59”	79°18’12”	By the fence of house in Dolon village.
	D22	50°39’53”	79°18’11”	By the fence of house in Dolon village.
	D23	50°39’50”	79°18’15”	By the fence of house in Dolon village.
	D24	50°40’38”	79°18’29”	At the entrance of Dolon village. Vegetation covering.
	D25	50°42’10”	79°17’50”	Plain near Dolon village.
Mostik	M1	50°40’56”	79° 6’18”	By the fence of house in Cheremushka village.
	M2	50°41’ 0”	79° 6’21”	Cemetery in Mostik village.
	M3	50°41’ 6”	79° 6’55”	By the fence of house in Mostik village.
	M4	50°41’19”	79° 6’44”	By the play-ground in Mostik village.
	M5	50°43’ 5”	79° 7’46”	Forest near Mostik village.
Cheremushka	CH1	50°38’44”	79° 3’37”	By the fence of house in Cheremushka village.
	CH2	50°38’43”	79° 3’39”	By the fence of house in Cheremushka village.
	CH3	50°38’43”	79° 3’38”	By the fence of house in Cheremushka village.
	CH4	50°38’39”	79° 3’39”	By the fence of house in Cheremushka village.
	CH5	50°38’37”	79° 3’38”	By the fence of house in Cheremushka village.
	CH6	50°38’37”	79° 3’34”	By the fence of house in Cheremushka village.
	CH7	50°38’37”	79° 3’29”	By the fence of house in Cheremushka village.
	CH8	50°38’35”	79° 3’19”	By the fence of house in Cheremushka village.
	CH9	50°38’42”	79° 3’ 2”	By the fence of house in Cheremushka village.
	CH10	50°38’43”	79° 3’ 9”	By the fence of house in Cheremushka village.
	CH11	50°38’44”	79° 3’18”	By the fence of house in Cheremushka village.
	CH12	50°38’45”	79° 3’30”	By the fence of house in Cheremushka village.
	CH13	50°38’44”	79° 3’38”	At the entrance of Cheremushka village (garden of ruined house).
	CH14	50°38’42”	79° 2’45”	Plain near Cheremushka (metalware found at 20 cm depth).
	CH15	50°38’34”	79° 3’ 3”	Near the river side of Irtysh (Cheremushka). Very hard ground.
Budene	B1	50°37’38”	79° 6’26”	Side of river in Budene village (submerged with flood).
	B2	50°37’38”	79° 6’26”	Side of river in Budene village (submerged with flood).
	B3	50°37’32”	79° 6’25”	Side of river in Budene village (submerged with flood).
	B4	50°37’32”	79° 6’25”	Side of river in Budene village (submerged with flood).
	B5	50°37’28”	79° 6’25”	Side of river in Budene village (submerged with flood).
	B6	50°37’21”	79° 6’19”	Drift near the fence in Budene village.
	B7	50°37’15”	79° 6’23”	End of Budene village.
	B8	50°37’17”	79° 6’29”	By the fence of house in Budene village.
	B9	50°37’12”	79° 6’33”	By the fence of ruined house in Budene village.
	B10	50°37’6”	79° 6’38”	Streetside in Budene village.
	B11	50°36’54”	79° 6’25”	By the toilet in Budene village.
	B12	50°36’57”	79° 6’37”	By the fence of house in Budene village.
	B13	50°36’51”	79° 6’24”	Plain near Budene village.

Radioactive measurements

Aliquots (70–100 g) of the soil samples dried were packed into a plastic vessel with a diameter of 6 cm and height of 2 cm, and γ-emitting radionuclides such as ^{137}Cs were determined by γ-ray spectrometry using an ordinary Ge and/or a planar type Ge detector. The spectrometers were calibrated with standards prepared from the New Brunswick Laboratory (NBL) reference materials No. 42-1 (4.04% uranium) and analytical grade KCl.

After γ-ray spectrometry, Pu analysis in soil was carried out using a sequential leaching method to find how the Pu from the local fallout debris was geochemically associated with soil. The $^{239,240}Pu$ determination was done for soil samples from sites in which higher levels and/or characteristic depth profiles of ^{137}Cs were found. Aliquots (30–40 g) of the samples were calcinated at about 500°C overnight and Pu was twice leached with concentrated HNO_3 with an addition of a small amount of H_2O_2 by heating at least for 3–4 hours (soluble fraction). Next, the residue obtained by centrifugation was decomposed with HNO_3 + HF + $HClO_4$ (residual fraction). After adding ^{242}Pu to each fraction as a tracer, Pu was separated and purified using an anion-exchange column method described by Yamamoto *et al.*[12,13] The purified Pu was electroplated onto a polished stainless steel disc, and its α-ray activities were determined by α-ray spectrometry. Accuracy and precision of the Pu analytical method used here was tested using some reference materials and shown to be satisfactory within 1–3% error.

An autoradiographic technique using CR-39 plastic α-track detector (BARYOTRAK-P) was applied to check the presence of hot-particles in each soil size fraction obtained by particle size discrimination. The samples (ca. 2 g) from each fraction were suspended with distilled water, and filtered with No. 5A filter paper to make a thin layer of soil. Then, the filter paper was gently dried at ca. 40°C and the detector (2 × 4 cm) was put on the filter paper and fixed with a tape. After standing for ca. 10 days under dark and cool condition, the detector was pre-etched in 6.5 M NaOH solution at 70 ± 1°C for 6 hours. The α-tracks on the detector were observed using a microscope.

RESULTS AND DISCUSSION

Accumulated levels of ^{137}Cs and $^{239,240}Pu$

The results of ^{137}Cs and $^{239,240}Pu$ measurements for the short (30 cm in depth) and long (100 cm in depth) soil cores are listed in Tables 2 and 3, respectively. The $^{239,240}Pu$ data are presented separately with both Pu fraction (soluble) leached by hot conc. $HNO_3 + H_2O_2$ and Pu fraction (residual) decomposed by HNO_3 + HF + $HClO_4$. All data are shown in terms of both activity per unit soil weight (Bq/kg) and activity per unit area (inventory, Bq/m^2).

As seen from Tables 2 and 3, for the 30 cm short cores, the ^{137}Cs and $^{239,240}Pu$ were found in layers up to a depth of

Table 2. $^{239,240}Pu$ and ^{137}Cs contents and inventories in soil from Dolon, Mostik, Cheremushka and Budene villages

No.	Depth (cm)	Concentration			
		^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
D1	0-5	7.54 ± 0.31	(344)	n.a.	
	5-10	5.10 ± 0.29	(280)	n.a.	
	10-15	2.01 ± 0.10	(136)	n.a.	
	15-20	0.38 ± 0.10	(28)	n.a.	
	20-25	n.d.		n.a.	
	Total		**(788)**		
D3	0-10	7.60 ± 0.30	(1040)	3.75 ± 0.12	(513)
	10-20	n.d.		0.10 ± 0.01	(16)
	20-30	n.d.		n.d.	
	30-40	n.d.		n.d.	
	40-50	n.d.		n.d.	
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-100	n.d.		n.d.	
	Total		**(1040)**		**(528)**
D4	0-10	14.29 ± 0.33	(1904)	3.45 ± 0.08	(558)
	10-20	n.d.		0.02 ± 0.01	(9)
	20-30	n.d.		n.d.	
	30-40	n.d.		n.d.	
	40-50	n.d.		n.d.	
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-100	n.d.		n.d.	
	Total		**(1904)**		**(567)**
D5	0-5	6.06 ± 0.44	(192)	3.15 ± 0.09	(100)
	5-10	15.25 ± 0.63	(540)	7.67 ± 0.34	(272)
	10-15	24.04 ± 0.67	(1141)	7.93 ± 0.34	(376)
	15-20	15.97 ± 0.65	(955)	10.09 ± 0.36	(604)
	20-28	23.34 ± 0.63	(2343)	20.36 ± 0.46	(2043)
	Total		**(5171)**		**(3395)**
D6	0-5	17.51 ± 0.54	(483)	n.a.	
	5-10	15.92 ± 0.47	(869)	n.a.	
	10-15	0.84 ± 0.13	(62)	n.a.	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(1413)**		
D7	0-5	7.98 ± 0.28	(496)	n.a.	
	5-10	14.31 ± 0.44	(958)	n.a.	
	10-15	22.54 ± 0.46	(1415)	n.a.	
	15-20	21.29 ± 0.39	(1551)	n.a.	
	20-29	8.66 ± 0.31	(846)	n.a.	
	Total		**(2870)**		
D8	0-5	4.18 ± 0.23	(217)	n.a.	
	5-10	5.30 ± 0.29	(339)	n.a.	
	10-15	2.86 ± 0.19	(200)	n.a.	
	15-20	4.92 ± 0.23	(371)	n.a.	
	20-30	4.58 ± 0.21	(853)	n.a.	
	Total		**(1980)**		

Table 2. continued

No.	Depth (cm)	Concentration ^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
D9	0-5	3.52 ± 0.17	(204)	n.a.	
	5-10	2.21 ± 0.20	(163)	n.a.	
	10-15	6.39 ± 0.26	(414)	n.a.	
	15-20	1.52 ± 0.17	(118)	n.a.	
	20-29	2.95 ± 0.23	(278)	n.a.	
	Total		**(1178)**		
D10	0-5	7.38 ± 0.30	(494)	1.86 ± 0.06	(125)
	5-10	9.36 ± 0.37	(694)	1.83 ± 0.07	(135)
	10-15	17.07 ± 0.24	(1043)	55.83 ± 1.40	(3410)
	15-20	21.76 ± 0.43	(1654)	13.72 ± 0.47	(1043)
	20-30	40.47 ± 0.73	(6428)	60.49 ± 1.69	(9608)
	Total		**(10314)**		**(14321)**
D11	0-5	10.84 ± 0.38	(541)	n.a.	
	5-10	12.85 ± 0.39	(782)	n.a.	
	10-15	15.17 ± 0.30	(1047)	n.a.	
	15-20	10.98 ± 0.39	(825)	n.a.	
	20-29	n.d		n.a.	
	Total		**(3196)**		
D12	0-5	2.79 ± 0.18	(143)	n.a.	
	5-10	8.22 ± 0.29	(647)	n.a.	
	10-15	24.16 ± 0.50	(1611)	n.a.	
	15-20	18.55 ± 0.47	(1599)	n.a.	
	20-29	1.98 ± 0.15	(319)	n.a.	
	Total		**(4319)**		
D13	0-5	28.67 ± 0.61	(1357)	30.91 ± 1.22	(1463)
	5-10	35.35 ± 0.70	(2118)	43.29 ± 1.14	(2593)
	10-15	25.89± 0.60	(1515)	34.17 ± 1.67	(1999)
	15-20	18.58 ± 0.53	(1150)	17.30 ± 0.61	(1070)
	20-30	n.d.		0.67 ± 0.03	(100)
	Total		**(6139)**		**(7226)**
D14	0-10	8.61 ± 0.29	(1356)	9.31 ± 0.14	(1465)
	10-20	0.19 ± 0.07	(32)	0.04 ± 0.00	(15)
	20-30	n.d.		0.02 ± 0.00	(6)
	30-40	n.d.		n.d.	
	40-50	n.d.		n.d.	
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-100	n.d.		n.d.	
	Total		**(1387)**		**(1486)**
D16	0-5	127.7 ± 1.16	(8005)	73.97 ± 3.59	(4637)
	5-10	7.33 ± 0.28	(494)	23.52 ± 0.53	(1586)
	10-15	2.13 ± 0.21	(172)	1.96 ± 0.07	(159)
	15-20	0.91 ± 0.12	(71)	0.41 ± 0.03	(32)
	20-30	n.d.		n.d.	
	Total		**(8742)**		**(6414)**
D17	0-5	128.2 ± 1.22	(7970)	n.a.	
	5-10	8.68 ± 0.36	(683)	n.a.	
	10-15	1.72 ± 0.17	(157)	n.a.	

Table 2. continued

No.	Depth (cm)	Concentration ^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(8811)**		
D19	0-5	11.66 ± 0.69	(447)	n.a.	
	5-10	7.91 ± 0.40	(662)	n.a.	
	10-15	17.89 ± 0.30	(1427)	n.a.	
	15-20	1.11 ± 0.16	(96)	n.a.	
	20-29	n.d.		n.a.	
	Total		**(2632)**		
D20	0-5	8.33 ± 0.69	(521)	21.23 ± 0.74	(1327)
	5-10	9.06 ± 0.32	(760)	6.68 ± 0.24	(560)
	10-15	9.5 ± 0.34	(818)	11.93 ± 0.37	(1027)
	15-20	18.68 ± 0.46	(1381)	31.43 ± 1.19	(2324)
	20-30	20.24 ± 0.39	(3169)	29.69 ± 0.98	(4650)
	Total		**(6649)**		**(9887)**
D21	0-5	3.48 ± 0.27	(168)	n.a.	
	5-10	7.21 ± 0.31	(495)	n.a.	
	10-15	3.62 ± 0.36	(248)	n.a.	
	15-20	0.86 ± 0.16	(63)	n.a.	
	20-30			n.d.	
	Total		**(974)**		
D22	0-5	13.7 ± 0.53	(524)	n.a.	
	5-10	20.54 ± 0.61	(869)	n.a.	
	10-15	5.19 ± 0.32	(287)	n.a.	
	15-20	4.19 ± 0.29	(198)	n.a.	
	20-25	5.52 ± 0.31	(473)	n.a.	
	Total		**(2351)**		
D23	0-5	6.41 ± 0.33	(418)	n.a.	
	5-10	6.11 ± 0.34	(460)	n.a.	
	10-15	4.73 ± 0.23	(315)	n.a.	
	15-20	3.57 ± 0.32	(249)	n.a.	
	20-30	2.56 ± 0.24	(451)	n.a.	
	Total		**(1894)**		
D24	0-10	71.35 ± 0.81	(9086)	94.92 ± 2.97	(12087)
	10-20	0.26 ± 0.07	(41)	0.11 ± 0.02	(11)
	20-30	n.d.		n.d.	
	30-40	n.d.		n.d.	
	40-50	n.d.		n.d.	
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-95	n.d.		n.d.	
	Total		**(9126)**		**(12097)**
D25	0-5	49.64 ± 0.89	(3185)	36.57 ± 1.11	(2347)
	5-10	11.97 ± 0.36	(896)	12.33 ± 0.38	(923)
	10-15	1.07 ± 0.17	(84)	0.31 ± 0.02	(24)
	15-20	n.d.		0.02 ± 0.01	(2)
	20-30	n.d.		n.d.	**(3296)**
	Total		**(4166)**		

Table 2. continued

No.	Depth (cm)	Concentration ^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
M1	0-5	5.86 ± 0.28	(481)	n.a.	
	5-10	1.18 ± 0.17	(103)	n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(583)**		
M2	0-5	16.28 ± 0.47	(1140)	2.8 ± 0.09	(196)
	5-10	4.16 ± 0.34	(224)	0.72 ± 0.03	(39)
	10-15	0.25 ± 0.08	(22)	0.04 ± 0.01	(4)
	15-20	n.d.		0.06 ± 0.01	(6)
	20-30	n.d.		n.d.	
	Total		**(1387)**		**(245)**
M3	0-5	4.45 ± 0.28	(321)	n.a.	
	5-10	7.82 ± 0.53	(468)	n.a.	
	10-15	5.93 ± 0.31	(539)	n.a.	
	15-20	1.82 ± 0.18	(135)	n.a.	
	20-30	2.41 ± 0.21	(297)	n.a.	
	Total		**(1761)**		
M4	0-5	9.99 ± 0.30	(533)	n.a.	
	5-10	9.01 ± 0.26	(562)	n.a.	
	10-15	5.27 ± 0.32	(459)	n.a.	
	15-20	0.74 ± 0.09	(73)	n.a.	
	20-30	n.d.		n.a.	
	Total		**(1627)**		
M5	0-5	59.62 ± 1.23	(2706)	2.76 ± 0.09	(125)
	5-10	0.93 ± 0.17	(61)	0.12 ± 0.02	(8)
	10-15	n.d.		n.d.	(3)
	15-20	n.d.		n.d.	
	20-30	n.d.		n.d.	
	Total		**(2767)**		**(137)**
CH1	0-5	10.04 ± 0.41	(752)	n.a.	
	5-10	8.80 ± 0.50	(540)	n.a.	
	10-15	5.11 ± 0.20	(387)	n.a.	
	15-20	7.17 ± 0.35	(485)	n.a.	
	20-28	0.08 ± 0.02	(13)	n.a.	
	Total		**(2177)**		
CH2	0-5	12.69 ± 0.24	(809)	n.a.	
	5-10	9.61 ± 0.33	(667)	n.a.	
	10-15	7.63 ± 0.25	(606)	n.a.	
	15-20	5.93 ± 0.27	(456)	n.a.	
	20-28	7.92 ± 0.39	(1079)	n.a.	
	Total		**(3616)**		
CH3	0-5	8.29 ± 0.40	(410)	n.a.	
	5-10	8.11 ± 0.30	(391)	n.a.	
	10-15	3.12 ± 0.22	(195)	n.a.	
	15-20	6.58 ± 0.33	(333)	n.a.	
	20-28	4.33 ± 0.24	(566)	n.a.	
	Total		**(1895)**		

Table 2. continued

No.	Depth (cm)	Concentration ^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
CH4	0-5	26.37 ± 0.64	(1234)	n.a.	
	5-10	22.36 ± 0.48	(1291)	n.a.	
	10-15	7.24 ± 0.22	(342)	n.a.	
	15-20	1.17 ± 0.15	(85)	n.a.	
	20-28	n.d.		n.a.	
	Total		**(2951)**		
CH5	0-5	12.78 ± 0.31	(756)	10.86 ± 0.38	(642)
	5-10	18.09 ± 0.66	(845)	3.33 ± 0.10	(155)
	10-15	23.01 ± 0.58	(1458)	37.27 ± 1.16	(2361)
	15-20	12.81 ± 0.33	(764)	2.59 ± 0.08	(155)
	20-29	15.27 ± 0.42	(1936)	46.2 ± 1.66	(5857)
	Total		**(5758)**		**(9170)**
CH6	0-5	7.1 ± 0.35	(408)	n.a.	
	5-10	3.84 ± 0.34	(252)	n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-29	n.d.		n.a.	
	Total		**(660)**		
CH7	0-5	16.39 ± 0.44	(1056)	n.a.	
	5-10	9.51 ± 0.26	(709)	n.a.	
	10-15	6.67 ± 0.21	(580)	n.a.	
	15-20	12.28 ± 0.41	(557)	n.a.	
	20-28	17.01 ± 0.41	(977)	n.a.	
	Total		**(3879)**		
CH8	0-5	9.54 ± 0.33	(622)	n.a.	
	5-10	4.17 ± 0.28	(330)	n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(952)**		
CH9	0-5	8.19 ± 0.28	(529)	n.a.	
	5-10	7.29 ± 0.34	(350)	n.a.	
	10-15	5.08 ± 0.21	(355)	n.a.	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(1234)**		
CH11	0-5	8.31 ± 0.30	(343)	n.a.	
	5-10	6.53 ± 0.34	(321)	n.a.	
	10-15	9.53 ± 0.25	(791)	n.a.	
	15-20	n.d.		n.a.	
	20-29	n.d.		n.a.	
	Total		**(1456)**		
CH12	0-5	16.01 ± 0.42	(1022)	1.46 ± 0.06	(93)
	5-10	28.07 ± 0.47	(1332)	3.02 ± 0.09	(143)
	10-15	19.42 ± 0.54	(1143)	20.28 ± 0.81	(1193)
	15-20	25.04 ± 0.46	(2072)	29.09 ± 1.09	(2407)
	20-30	5.80 ± 0.46	(705)	1.37 ± 0.06	(167)
	Total		**(6274)**		**(4003)**

Table 2. continued

No.	Depth (cm)	Concentration			
		^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
CH13	0-10	10.27 ± 0.46	(1155)	0.99 ± 0.04	(111)
	10-20	14.47 ± 0.48	(2028)	14.76 ± 0.45	(2070)
	20-30	19.63 ± 0.46	(2491)	71.62 ± 1.69	(9091)
	30-40	4.36 ± 0.17	(515)	13.92 ± 0.40	(1645)
	40-50	n.d.		0.03 ± 0.01	(3.4)
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-95	n.d.		n.d.	
	Total		**(6189)**		**(12920)**
CH14	0-10	15.11 ± 0.42	(1813)	49.91 ± 1.46	(5988)
	10-20	46.39 ± 0.87	(6351)	22.96 ± 0.67	(3144)
	20-30	1.03 ± 0.15	(128)	0.56 ± 0.03	(70)
	30-40	0.37 ± 0.12	(46)	0.11 ± 0.01	(14)
	40-50	0.18 ± 0.06	(20)	0.08 ± 0.01	(9)
	50-60	n.d.		n.d.	
	60-80	n.d.		n.d.	
	80-100	n.d.		n.d.	
	Total		**(8357)**		**(9225)**
CH15	0-10	5.39 ± 0.24	(793)	n.a.	
	10-20	0.29 ± 0.06	(37)	n.a.	
	20-30	0.04 ± 0.01	(6)	n.a.	
	30-40	n.d.		n.a.	
	40-50	n.d.		n.a.	
	Total		**(837)**		
B1	0-5	14.64 ± 0.68	(800)	10.23 ± 0.31	(559)
	5-10	9.12 ± 0.45	(643)	6.21 ± 0.17	(438)
	10-15	5.63 ± 0.29	(348)	10.53 ± 0.27	(651)
	15-20	1.97 ± 0.21	(164)	11.32 ± 0.38	(944)
	20-29	0.9 ± 0.11	(135)	0.22 ± 0.01	(33)
	Total		**(2091)**		**(2625)**
B3	0-5	16.39 ± 0.40	(867)	n.a.	
	5-10	14.07 ± 1.03	(660)	n.a.	
	10-15	11.61 ± 1.20	(847)	n.a.	
	15-20	8.71 ± 0.44	(609)	n.a.	
	20-29	7.43 ± 0.45	(1186)	n.a.	
	Total		**(4169)**		
B4	5-10	7.71 ± 0.35	(551)	11.43 ± 0.35	(817)
	10-15	4.94 ± 0.20	(343)	4.14 ± 0.12	(288)
	15-20	4.69 ± 0.31	(309)	1.69 ± 0.07	(111)
	20-30	2.42 ± 0.15	(353)	1.26 ± 0.05	(184)
	Total		**(2566)**		**(2929)**
B5	0-5	8.68 ± 0.64	(547)	n.a.	
	5-10	3.00 ± 0.16	(196)	n.a.	
	10-15	2.12 ± 0.36	(157)	n.a.	
	15-20	1.97 ± 0.35	(113)	n.a.	
	20-30	0.45 ± 0.11	(83)	n.a.	
	Total		**(1097)**		
B6	0-5	3.72 ± 0.22	(261)	n.a.	
	5-10	n.d.		n.a.	

Table 2. continued

No.	Depth (cm)	Concentration			
		^{137}Cs (Bq/kg dry)		$^{239,240}Pu$ (Bq/kg dry)	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-29	n.d.		n.a.	
	Total		**(261)**		
B7	0-5	26.1 ± 1.26	(1229)	3.88 ± 0.15	(182)
	5-10	67.03 ± 1.09	(4176)	34.04 ± 1.06	(2121)
	10-15	16.62 ± 0.72	(1340)	16.74 ± 0.58	(1349)
	15-20	1.95 ± 0.20	(122)	1.77 ± 0.06	(111)
	20-29	0.75 ± 0.09	(112)	0.03 ± 0.01	(4)
	Total		**(6979)**		**(3767)**
B8	0-5	9.75 ± 0.39	(343)	1.88 ± 0.06	(66)
	5-10	9.07 ± 0.44	(667)	1.56 ± 0.03	(115)
	10-15	10.48 ± 0.34	(757)	2.31 ± 0.09	(167)
	15-20	7.42 ± 0.30	(464)	1.29 ± 0.03	(80)
	20-30	5.91 ± 0.39	(973)	1.49 ± 0.04	(246)
	Total		**(3205)**		**(673)**
B9	5-10	n.d.		n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-30	1.6 ± 0.16	(277)	n.a.	
	Total		**(277)**		
B10	0-5	3.81 ± 0.24	(276)	4.39 ± 0.10	(318)
	5-10	4.73 ± 0.23	(334)	0.19 ± 0.01	(14)
	10-15	2.7 ± 0.17	(228)	0.21 ± 0.01	(18)
	15-20	2.27 ± 0.16	(210)	0.52 ± 0.03	(v48)
	20-30	5.85 ± 0.24	(926)	5.41 ± 0.1	(856)
	Total		**(1974)**		**(1254)**
B11	0-5	5.44 ± 0.38	(341)	n.a.	
	5-10	3.05 ± 0.30	(193)	n.a.	
	10-15	5.02 ± 0.35	(450)	n.a.	
	15-20	6.57 ± 0.37	(540)	n.a.	
	20-30	n.d.		n.a.	
	Total		**(1525)**		
B12	0-5	2.59 ± 0.23	(136)	n.a.	
	5-10	n.d.		n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-30	n.d.		n.a.	
	Total		**(136)**		
B13	0-5	8.22 ± 0.48	(600)	n.a.	
	5-10	0.18 ± 0.06	(15)	n.a.	
	10-15	n.d.		n.a.	
	15-20	n.d.		n.a.	
	20-29	n.d.		n.a.	
	Total		**(615)**		

Parenthesis shows the inventory of the activity (Bq/m^2).
Error shows 1σ standard deviation from counting statistics.
n.d.: not detected.
n.a.: not analyzed.

Table 3. Results of stepwise leaching of $^{239,240}Pu$ in soil from Dolon, Mostik, Cheremushka and Budene villages.

No.	Depth (cm)	Soluble Pu $^{239,240}Pu$ (Bq/kg dry)	Soluble Pu ^{238}Pu/$^{239,240}Pu$	Soluble Pu $^{239,240}Pu$ (Bq/m^2)	Residual Pu $^{239,240}Pu$ (Bq/kg dry)	Residual Pu ^{238}Pu/$^{239,240}Pu$	Residual Pu $^{239,240}Pu$ (Bq/m^2)	Total $^{239,240}Pu$ (Bq/m^2)	Soluble $^{239,240}Pu$ (%)
D3	0-10	0.35 ± 0.02		47.5	3.40 ± 0.12	0.046 ± 0.004	465.2	512.7	9.3
	10-20	n.d.			0.10 ± 0.01		15.5	15.5	
	20-30	n.d.			n.d.				
	30-40	n.d.			n.d.				
	40-50	n.d.			n.d.				
	50-60	n.d.			n.d.				
	60-80	n.d.			n.d.				
	80-100	n.d.			n.d.				
	Total			**47.5**			**480.8**	**528.3**	**9.0**
D4	0-10	0.75 ± 0.04	0.053 ± 0.207	100.2	2.68 ± 0.07	0.037 ± 0.003	357.6	457.8	21.9
	10-20	0.02 ± 0.01		3.7	0.01 ± 0.00		1.8	5.5	66.9
	20-30	n.d.			n.d.				
	30-40	n.d.			n.d.				
	40-50	n.d.			n.d.				
	50-60	n.d.			n.d.				
	60-80	n.d.			n.d.				
	80-100	n.d.			n.d.				
	Total			**103.9**			**359.4**	**463.3**	**22.4**
D5	0-5	0.93 ± 0.04	0.029 ± 0.194	29.6	0.08 ± 0.01	0.027 ± 0.004	70.4	100.0	29.6
	5-10	1.12 ± 0.05	0.034 ± 0.195	39.6	0.33 ± 0.11	0.039 ± 0.004	232.0	271.6	14.6
	10-15	1.01 ± 0.06	0.043 ± 0.227	47.9	0.33 ± 0.11	0.038 ± 0.004	328.2	376.1	12.7
	15-20	1.12 ± 0.06	0.039 ± 0.192	66.8	0.36 ± 0.13	0.028 ± 0.003	537.2	604.0	11.1
	20-28	1.64 ± 0.04	0.037 ± 0.096	164.8	0.45 ± 0.21	0.038 ± 0.001	1878.4	2043.3	8.1
	Total			**348.8**			**3046.2**	**3394.9**	**10.3**
D10	0-5	0.27 ± 0.02		17.9	1.60 ± 0.06	0.023 ± 0.004	107.0	124.9	14.3
	5-10	0.40 ± 0.02		29.7	1.43 ± 0.07	0.026 ± 0.006	105.8	135.4	21.9
	10-15	33.74 ± 1.11	0.012 ± 0.070	2060.9	22.09 ± 0.85	0.037 ± 0.002	1349.1	3410.0	60.4
	15-20	1.50 ± 0.05	0.049 ± 0.112	114.1	12.22 ± 0.47	0.030 ± 0.002	928.8	1042.9	10.9
	20-30	5.12 ± 0.20	0.037 ± 0.099	813.6	55.36 ± 1.68	0.033 ± 0.001	8794.3	9607.8	8.5
	Total			**3036.1**			**11284.9**	**14321.0**	**21.2**
D13	0-5	2.33 ± 0.10	0.040 ± 0.139	110.3	28.58 ± 1.22	0.033 ± 0.002	1352.7	1463.0	7.5
	5-10	13.78 ± 0.43	0.042 ± 0.056	825.3	29.51 ± 1.06	0.036 ± 0.002	1768.0	2593.3	31.8
	10-15	2.30 ± 0.09	0.041 ± 0.130	134.6	31.87 ± 1.67	0.036 ± 0.002	1864.4	1999.0	6.7
	15-20	2.07 ± 0.09	0.037 ± 0.145	128.1	15.23 ± 0.60	0.034 ± 0.002	942.1	1070.1	12.0
	20-30	0.11 ± 0.01		17.2	0.55 ± 0.03		83.0	100.2	17.2
	Total			**1215.4**			**6010.1**	**7225.5**	**16.8**
D14	0-10	4.70 ± 0.1	0.046 ± 0.064	739.8	4.61 ± 0.07	0.043 ± 0.002	725.6	1465.4	50.5
	10-20	n.d.			0.04 ± 0.00		7.3	7.3	
	20-30	n.d.			0.02 ± 0.00		2.9	2.9	
	30-40	n.d.			n.d.				
	40-50	n.d.			n.d.				
	50-60	n.d.			n.d.				
	60-80	n.d.			n.d.				
	80-100	n.d.			n.d.				
	Total			**739.8**			**735.7**	**1475.5**	**50.1**
D16	0-5	17.05 ± 0.48	0.038 ± 0.049	1068.8	56.92 ± 3.56	0.033 ± 0.002	3568.3	4637.1	23.0
	5-10	12.05 ± 0.43	0.041 ± 0.067	812.4	11.48 ± 0.31	0.041 ± 0.002	774.0	1586.5	51.2

	10-15	0.34 ± 0.03		27.2	1.62 ± 0.06	0.050 ± 0.006	131.5	158.7	17.2
	15-20	0.11 ± 0.01		8.3	0.30 ± 0.02		23.3	31.7	26.3
	20-30	n.d.			n.d.				
	Total			**1916.8**			**4497.1**	**6413.9**	**29.9**
D20	0-5	0.25 ± 0.02		15.8	20.98 ± 0.74	0.037 ± 0.002	1310.9	1326.7	1.2
	5-10	0.46 ± 0.02	0.031 ± 0.244	38.8	6.22 ± 0.24	0.032 ± 0.003	521.5	560.3	6.9
	10-15	0.24 ± 0.01	0.072 ± 0.214	20.3	11.69 ± 0.37	0.038 ± 0.002	1006.8	1027.1	2.0
	15-20	1.07 ± 0.04	0.024 ± 0.168	78.9	30.37 ± 1.19	0.040 ± 0.002	2244.8	2323.7	3.4
	20-30	0.71 ± 0.03	0.020 ± 0.237	111.7	28.98 ± 0.98	0.040 ± 0.002	4538.0	4649.7	2.4
	Total			**265.5**			**9622.0**	**9887.5**	**2.7**
D24	0-10	18.42 ± 0.42	0.028 ± 0.052	2345.0	76.50 ± 2.94	0.038 ± 0.001	9741.5	12086.5	19.4
	10-20	0.01 ± 0.00		1.8	0.06 ± 0.01		8.8	10.6	16.9
	20-30	n.d.			0.03 ± 0.01		4.8	4.8	
	30-40	n.d.			n.d.				
	40-50	n.d.			n.d.				
	50-60	n.d.			n.d.				
	60-80	n.d.			n.d.				
	80-95	n.d.			n.d.				
	Total			**2346.8**			**9755.2**	**12101.9**	**19.4**
D25	0-5	5.19 ± 0.25	0.033 ± 0.134	332.9	31.38 ± 1.09	0.038 ± 0.001	2013.8	2346.7	14.2
	5-10	1.59 ± 0.07	0.041 ± 0.149	118.9	10.74 ± 0.37	0.040 ± 0.002	803.9	922.8	12.9
	10-15	0.08 ± 0.01		6.6	0.22 ± 0.01		17.5	24.1	27.3
	15-20	0.02 ± 0.01		1.9	n.d.			1.9	
	20-30	n.d.			n.d.				
	Total			**460.3**			**2835.3**	**3295.5**	**14.0**
M2	0-5	1.09 ± 0.03		76.3	1.71 ± 0.09	0.039 ± 0.007	119.7	196.0	38.9
	5-10	0.20 ± 0.01		10.7	0.52 ± 0.03		28.2	39.0	27.6
	10-15	0.014 ± 0.003		1.26	0.031 ± 0.005		2.8	4.0	31.1
	15-20	n.d.			0.063 ± 0.009		6.3	6.3	
	20-30	n.d.			n.d.				
	Total			**88.3**			**157.0**	**245.3**	**36.0**
M5	0-5	0.049 ± 0.008	0.038 ± 0.004	2.21	2.71 ± 0.09	0.036 ± 0.004	122.9	125.1	1.8
	5-10	0.087 ± 0.016	0.035 ± 0.004	5.75	0.037 ± 0.007		2.4	8.2	70.1
	10-15	0.039 ± 0.009	0.042 ± 0.007	3.39	n.d.			3.4	
	15-20	n.d.			n.d.				
	20-30	n.d.			n.d.				
	Total			**11.4**			**125.3**	**136.7**	**8.3**
CH5	0-5	0.79 ± 0.03	0.042 ± 0.007	46.8	10.07 ± 0.38	0.041 ± 0.003	595.3	642.1	7.3
	5-10	1.15 ± 0.05	0.037 ± 0.007	53.8	2.18 ± 0.09	0.033 ± 0.005	101.6	155.4	34.6
	10-15	2.58 ± 0.09	0.040 ± 0.004	163.6	34.69 ± 1.16	0.039 ± 0.001	2197.7	2361.3	6.9
	15-20	0.64 ± 0.03	0.044 ± 0.010	38.3	1.95 ± 0.08	0.035 ± 0.005	116.4	154.7	24.7
	20-29	1.38 ± 0.06	0.036 ± 0.006	175.0	44.82 ± 1.66	0.037 ± 0.001	5681.8	5856.7	3.0
	Total			**477.4**			**8692.8**	**9170.2**	**5.2**
CH12	0-5	0.47 ± 0.02	0.040 ± 0.010	30.1	0.99 ± 0.05	0.035 ± 0.008	63.1	93.2	32.3
	5-10	0.98 ± 0.04	0.042 ± 0.008	46.7	2.03 ± 0.08	0.036 ± 0.005	96.5	143.1	32.6
	10-15	0.56 ± 0.02	0.035 ± 0.006	33.2	19.71 ± 0.81	0.039 ± 0.002	1160.1	1193.3	2.8
	15-20	7.71 ± 0.29	0.039 ± 0.003	638.0	21.38 ± 1.05	0.021 ± 0.002	1768.6	2406.6	26.5
	20-30	0.24 ± 0.01	0.043 ± 0.008	28.9	1.14 ± 0.06	0.044 ± 0.009	138.2	167.2	17.3
	Total			**776.9**			**3226.5**	**4003.4**	**19.4**

CH13	0-10	0.28 ± 0.01	0.042 ± 0.009	31.6	0.71 ± 0.04	0.041 ± 0.009	79.7	111.3	28.4
	10-20	0.51 ± 0.02	0.038 ± 0.008	72.0	14.25 ± 0.45	0.039 ± 0.002	1997.6	2069.6	3.5
	20-30	19.75 ± 0.58	0.040 ± 0.002	2506.8	51.87 ± 1.59	0.041 ± 0.001	6583.9	9090.7	27.6
	30-40	0.014 ± 0.003		1.7	13.91 ± 0.40	0.040 ± 0.002	1643.0	1644.7	0.1
	40-50	0.005 ± 0.002		0.6	0.02 ± 0.01		2.8	3.4	18.2
	50-60	n.a.			n.a.				
	60-80	n.a.			n.a.				
	80-95	n.a.			n.a.				
	Total			**2613**			**10307**	**12920**	**20.2**
CH14	0-10	0.63 ± 0.02	0.038 ± 0.006	75.3	49.28 ± 1.46	0.041 ± 0.001	5913.1	5988.4	1.3
	10-20	2.95 ± 0.07	0.035 ± 0.003	403.8	20.01 ± 0.67	0.039 ± 0.002	2739.8	3143.6	12.8
	20-30	0.48 ± 0.03		59.5	0.084 ± 0.008		10.5	70.0	85.0
	30-40	0.076 ± 0.008		9.3	0.035 ± 0.008		4.4	13.7	68.1
	40-50	0.052 ± 0.008		5.8	0.032 ± 0.003		3.5	9.3	62.1
	50-60	n.d.			n.d.				
	60-80	n.a.			n.a.				
	80-100	n.a.			n.a.				
	Total			**553.6**			**8671.3**	**9224.9**	**6.0**
B1	0-5	1.28 ± 0.04	0.042 ± 0.005	69.7	8.96 ± 0.31	0.031 ± 0.002	489.5	559.2	12.5
	5-10	0.81 ± 0.04	0.045 ± 0.008	57.4	5.40 ± 0.16	0.042 ± 0.003	380.4	437.8	13.1
	10-15	2.46 ± 0.07	0.043 ± 0.003	152.5	8.06 ± 0.27	0.038 ± 0.003	499.0	651.5	23.4
	15-20	0.20 ± 0.02		16.8	11.12 ± 0.38	0.032 ± 0.003	926.8	943.6	1.8
	20-29	0.06 ± 0.005		8.5	0.16 ± 0.01		24.1	32.5	26.0
	Total			**304.8**			**2319.9**	**2624.7**	**11.6**
B4	0-5	1.99 ± 0.05	0.040 ± 0.004	103.8	27.31 ± 0.79	0.037 ± 0.001	1426.3	1530.1	6.8
	5-10	1.26 ± 0.04	0.041 ± 0.004	90.0	10.17 ± 0.35	0.034 ± 0.002	726.5	816.5	11.0
	10-15	0.88 ± 0.04	0.042 ± 0.007	61.4	3.26 ± 0.11	0.037 ± 0.004	226.4	287.8	21.3
	15-20	0.82 ± 0.04	0.032 ± 0.007	54.1	0.87 ± 0.05		57.0	111.2	48.7
	20-30	0.42 ± 0.02		61.8	0.84 ± 0.05		121.7	183.6	33.7
	Total			**371.1**			**2558.0**	**2929.1**	**12.7**
B7	0-5	1.17 ± 0.05	0.038 ± 0.007	54.9	2.71 ± 0.14	0.039 ± 0.007	127.5	182.4	30.1
	5-10	10.37 ± 0.48	0.020 ± 0.002	646.3	23.67 ± 0.95	0.039 ± 0.002	1474.7	2121.0	30.5
	10-15	0.45 ± 0.02	0.034 ± 0.008	36.1	16.29 ± 0.58	0.045 ± 0.002	1312.8	1348.8	2.7
	15-20	0.036 ± 0.006		2.3	1.74 ± 0.06	0.047 ± 0.006	108.2	110.5	2.0
	20-29	0.025 ± 0.005		3.8	n.d.			3.8	
	Total			**743.4**			**3023.2**	**3766.6**	**19.7**
B8	0-5	0.46 ± 0.02		16.3	1.42 ± 0.05		49.9	66.2	24.6
	5-10	0.37 ± 0.02	0.040 ± 0.007	27.3	1.19 ± 0.03		87.2	114.6	23.8
	10-15	0.36 ± 0.01	0.043 ± 0.006	25.9	1.95 ± 0.09		140.7	166.7	15.6
	15-20	0.66 ± 0.02	0.037 ± 0.005	41.2	0.63 ± 0.02	0.044 ± 0.006	39.2	80.4	51.2
	20-30	0.26 ± 0.01		43.2	1.23 ± 0.04	0.040 ± 0.005	202.4	245.6	17.6
	Total			**153.9**			**519.5**	**673.4**	**22.9**
B10	0-5	0.097 ± 0.007		7.0	4.29 ± 0.10	0.035 ± 0.003	311.2	318.2	2.2
	5-10	0.12 ± 0.01		8.1	0.076 ± 0.006		5.4	13.5	60.3
	10-15	0.10 ± 0.01		8.3	0.12 ± 0.01		9.7	18.1	46.1
	15-20	0.075 ± 0.005		6.9	0.45 ± 0.03		41.4	48.4	14.3
	20-30	2.02 ± 0.05	0.021 ± 0.003	319.8	3.39 ± 0.08	0.032 ± 0.002	535.8	855.6	37.4
	Total			**350.3**			**903.5**	**1253.7**	**27.9**

Error shows 1σ– standard deviation from counting statistics.
n.d.: not detected.
n.a.: not analyzed.

30 cm. These depth profiles are characterized by the following four features: (1) high inventory in surface layer, (2) gradual increase of inventory with depth, (3) subsurface maximum of inventory with depth, and (4) exponential decrease of inventory with depth. For the 100 cm cores from Dolon, more than 90% of the ^{137}Cs and ^{239,240}Pu contents were found within a depth of 10 cm.[7] For the long cores from Cheremushka, both nuclides were within a depth of ca. 30 cm. After considering many depth profiles of ^{137}Cs and ^{239,240}Pu contents previously obtained around the SNTS by Yamamoto *et al.*[2], we at first expected the occurrence of some ^{137}Cs and ^{239,240}Pu in the deeper layers up to a depth

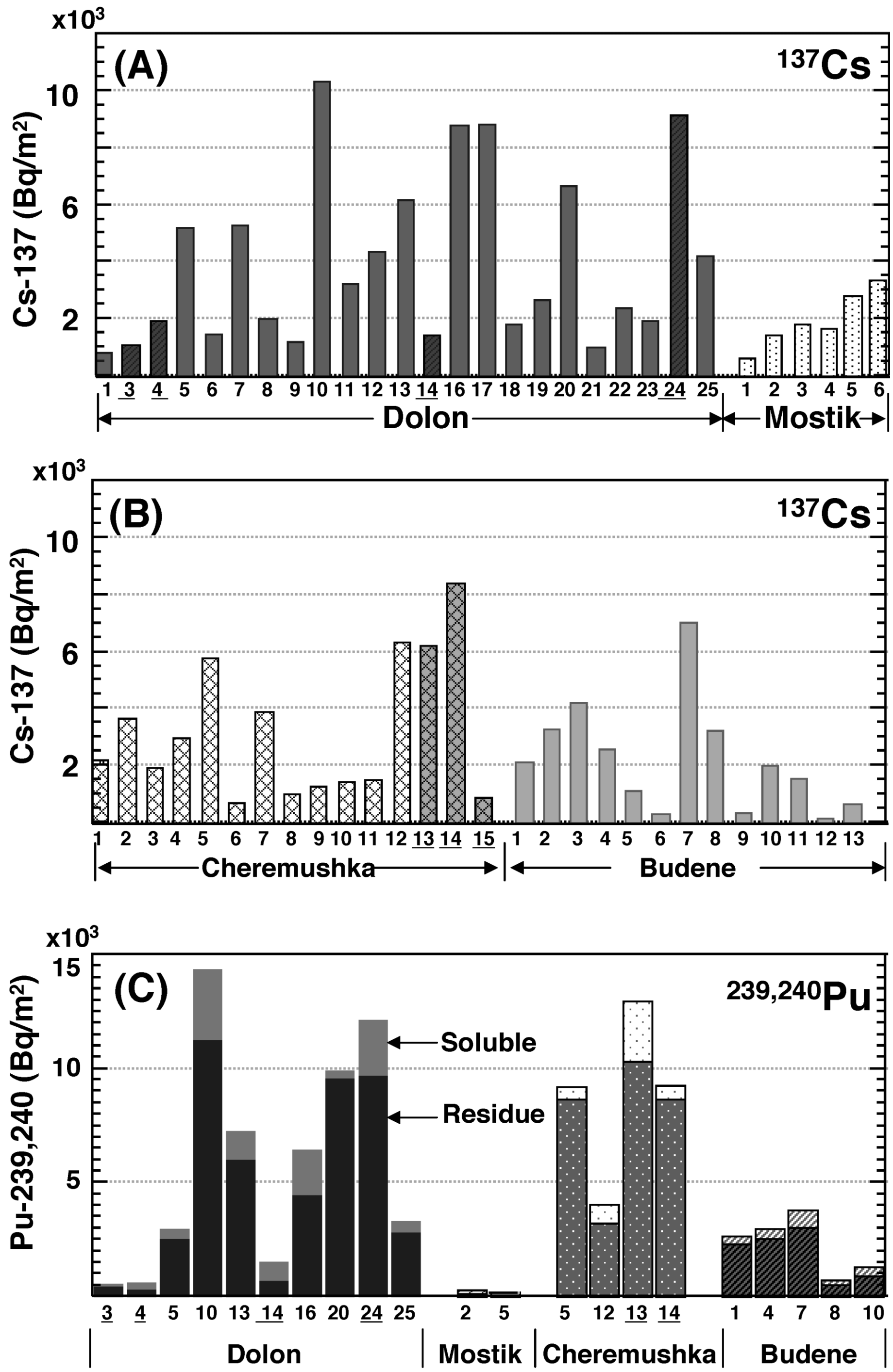

Fig. 2. Inventories of ^{239,240}Pu and ^{137}Cs in soils from Dolon, Mostik, Cheremushka and Budene settlements. Underlining of a number means the 100 cm soil core sample.

of 100 cm. But, such ^{137}Cs and $^{239,240}Pu$ were undetectable in all the long soil cores examined. The depth profiles of these nuclides depend largely on the sampling site. In fact, according to local inhabitants, the sites chosen for the 100 cm cores at Dolon village have not been disturbed or plowed for a long time. Usually, depth profiles of these nuclides in soil from the undisturbed areas are characterized by exponential decrease with depth. Within the villages, it is mostly the sampling at sample locations that is responsible for the artificial disturbance of soil. Furthermore, it must be kept in mind that prevailing strong wind and rain may promote the removal of surface soil and/or chemical and physical weathering of soil and as a result, deposited radionuclides will be redistributed.

Although there are high uncertainties due to the small surface area collected and the lack of soil sampling down to a sufficient depth at which both nuclides become less than detectable, it is very important to estimate the accumulated levels of ^{137}Cs and $^{239,240}Pu$ at this point of time. Comparison with the inventory (Fig. 2) seems to be more meaningful because activity per unit soil weight varies with a large number of parameters (soil type, topography, water content, mineralogy, rate of soil erosion, rainfall, *etc*). The estimated inventories of ^{137}Cs were found in the wide range of 140 - 10,310 Bq/m^2. The average values of ^{137}Cs estimated from the 30 cm short cores were as follows: 4090, 1910, 2690 and 2170 Bq/m^2 at Dolon, Mostik, Cheremushka and Budene villages, respectively. The $^{239,240}Pu$ inventories for all the villages were in the range from 140 - 14,310 Bq/m^2, and mean values for the 30 cm core samples were 7360, 190, 6590 and 2250 Bq/m^2 at Dolon, Mostik, Cheremushka and Budene villages, respectively. As a whole, the inventories of both nuclides show the presence of hotspot-like sites with a random deposition. The accumulated levels for $^{239,240}Pu$ found in Dolon and Cheremushka are clearly higher than those of Mostik and Budene. This seems to reflect the trace of the radioactive cloud from the first atomic bomb.

Until now, current ^{137}Cs and $^{239,240}Pu$ contamination levels on land from the various areas, including some settlements such as Chagan, Sarzhal, Kainar and Karaul[6,13] and the areas from Semipalatinsk City to Kurchatov City, and to Koresteli settlement north-east of the test site[13] and to the Ust'-Kamenogorsk City located more than 300 km east of the SNTS have been monitored by our missions.[14,15] The maximum level found in the village of Dolon belongs to the highest group among the inventories of both nuclides obtained at the above-mentioned areas. In the villages of Dolon and Mostik, the high variability of ^{137}Cs, ^{90}Sr and $^{239,240}Pu$ levels has been reported by some investigators (Table 4). Although the data obtained here are not greatly

Table 4 Comparisons of radioactivity accumulation levels in soil samples from Dolon and its adjoining settlements.

Author	Site	Core soil (cm)		Inventory in soil (Bq/m^2)			Concentration in surface soil (Bq/kg)				Ref.
		Diameter	Depth	^{137}Cs	$^{239,240}Pu$	^{90}Sr	^{137}Cs	$^{239,240}Pu$	^{90}Sr	^{241}Am	
Steinhausler, F. *et al*.	Dolon	8	0-12, -14	ca.500-ca. 8000	ca.2000-20800	ca.400-ca .600					11
Shebell, P. and Hutter, A.R.	Dolon	8.9	0-15	ca.10400			47.5*				10
Hille, R. *et al*.	Mostik		0-5				5-60		4-12	0.9-1.6	21
Gastberger, M. *et al*.	Dolon (-Mostik)	8	0-12, -14	300-7900		500-6100	<1-34		n.d.-26		8
Gastberger, M. *et al*.	Dolon (-Mostik)	8	0-12, -14		140-9700			0.7-41.6			9
Duvasov, Y. *et al*.	Dolon (-Mostik)				40-11000 (37000)**						22
Deriglasow, W.N. *et al*.	Dolon			5200-7900		1500-5800					23
Yamamoto, M. *et al*.	Dolon, Mostik	4.7	0-10,30	1480, 3440	2230, 380		4.5*	0.62*			6
This work	Dolon	4.7	0-30, -100***	790-10310	530-14320		2.8-71.4*	1.7-94.9*			7
	Mostik		0-30	580-2770	140, 250		4.5-59.7*	2.8*			
	Cheremushka		0-30, -100***	660-8360	4000-12920		5.4-26.4*	1.0-49.9*			
	Budene		0-30	140-6980	670-3770		0.6-26.1*	1.6-29.3*			

* 0-5 cm in depth.
** This value was found in sample collected between Dolon and Mostik.
*** Two cores (4.7 cm in diameter x2) were collected and mixed.

different from these data, it is worthy of note that ^{137}Cs and $^{239,240}Pu$ inventories over 1.0×10^4 Bq/m^2 are still observed at some sites within the villages even after about 50 years from the first nuclear weapon test. The current distributions of ^{137}Cs and $^{239,240}Pu$ are reflecting various factors such as migration into soil, washing out, resuspension, human activities and so on. It seems acceptable to assume that the maximum values in the distribution are more likely to reflect the original level of fallout contamination than the small values. According to the UNSCEAR report,[16] global fallout ^{137}Cs inventory is estimated to be about 500 Bq/m^2 in the latitude band of 40–50°. The average level at the Altai region, north of Semipalatinsk City, is also reported to be about 50 mCi/km^2 (1,850 Bq/m^2).[17]

The $^{239,240}Pu/^{137}Cs$ activity ratios for total inventories were in the range from 0.05–1.6, and similar values have been observed by Gastberger *et al*. (0.2–2.3 in soils of 12 cm in depth)[9] and by Yamamoto *et al*. (0.2–1.5 in soils of 30 cm in depth).[6] These values are several tens of times higher than the global fallout values of 0.02–0.03.[18] The higher $^{239,240}Pu/^{137}Cs$ activity ratios strongly indicate that more Pu deposited than ^{137}Cs by the local fallout from atomic explosions at the SNTS. The $^{240}Pu/^{239}Pu$ atomic ratios in soil from these area are reported to be 0.03–0.05 by Yamamoto *et al.*[6] It is clear that the deposited Pu around here is, therefore, attributed to the weapons-grade Pu from the atomic bombs. In contrast, the $^{238}Pu/^{239,240}Pu$ activity ratios were in the narrow range of 0.03–0.05, which were nearly the same as the values (0.03–0.04) found in global fallout in the northern Hemisphere.[18]

Characteristics of ^{137}Cs and Pu in soil

To obtain insight into the characteristics of Pu deposited in soil, a stepwise leaching of Pu from soil was examined (Table 3). For soil contaminated with global fallout from atmospheric weapons tests, Pu has been recognized to be almost quantitatively leached by digesting with conc. HNO_3 + H_2O_2. As can be seen from Table 3, 15% on average (ranging from 3–50%) is soluble and most Pu remains in the soil residue. These results are almost the same as the results obtained previously.[6] A small portion of Pu found in the soluble fraction may be attributable to the global fallout Pu and loosely bound- local Pu from the SNTS. We do not know whether this soluble Pu fraction becomes mobile or not under these dry environmental conditions. Most of the Pu is also clearly incorporated into various sizes of fused particles formed in the course of the condensation of melting materials such as vaporized soil and bomb materials. This result may elucidate the rather low $^{239,240}Pu$ concentrations that have been found in biota and foodstuffs around the SNTS.[8,11,14] The detailed observation of both soluble and residual Pu depth profiles from the undisturbed areas indicates that most of the Pu found below the surface soil layer might be transported predominantly by downward migration of fused small size particles including Pu. The Pu downward in this area is, therefore, predicted to be very low unless soil layers are disturbed. Generally, the Pu percentage in the residual soil fraction shows a tendency to decrease with increasing distance from the SNTS.[15]

Furthermore, particle size discrimination was undertaken by using the surface soil (site D24: 10 cm in depth) from Dolon settlement. The particle size dependencies of the distribution of ^{137}Cs and $^{239,240}Pu$ are interesting from the viewpoints of the inhalation of resuspended materials and/or migration processes to deeper soil layer.

The results are shown in Fig. 3. As seen from this figure, the concentrations of ^{137}Cs and $^{239,240}Pu$ increase apparently with the decrease of soil particle size. There are differences of the both nuclide contents in particle size fractions between >125 μm and <125 μm, that is, concentrations in the 4 fractions of size <125 μm are several times higher than those in the 3 fractions of size >125 μm. In the fractions smaller than 125 μm, the percentage contributions of soil weight and $^{239,240}Pu$ and ^{137}Cs contents were 24.8, 56.2 and 60.5% of the total, respectively. In all the fractions, more than 90% of $^{239,240}Pu$ was not leached with conc.HNO_3 + H_2O_2, and the $^{238}Pu/^{239,240}Pu$ activity ratios were 0.033–0.063 for the soluble fractions and 0.036–0.041 for the residual fractions. The $^{239,240}Pu/^{137}Cs$ activity ratios, except for the 2,000–500 μm size fraction, did not change so greatly, ranging from 1.0 to 1.5, indicating that fractionation between Pu and ^{137}Cs does not happen to a large extent. Kazachevski *et al.*[19] also reported the particle size dependence of radionuclides in surface soil samples (0–5 cm) collected in the SNTS territory. The highest concentrations were found in the 400–500 μm fraction for ^{90}Sr, ^{137}Cs and ^{241}Am, and 500–1000 μm fraction for $^{239,240}Pu$, and the percentage contributions of their contents were the largest in the 500–1000 mm fractions. These results, including our results, indicate that within the test site heavy and larger fused particles including high contents of radionuclides are preferentially deposited, and in the outside, relatively smaller size particles are responsible for the contamination of radionuclides on land.

An autoradiographic technique using the CR-39 plastic α-track detector was also applied to confirm whether hot-particles of Pu were present or absent in the soil. If α-particles are emitted from sub-micron size Pu oxide particles or from a conglomerates of such particles, a "cluster or star" is formed in the detecting media. Although a quantitative analysis, including the chemical form and effective density of the Pu, was not performed this time for each soil fraction, the most striking feature is that a star-like pattern of α-tracks, probably from Pu, was detected clearly as shown in Fig. 4. These stars were abundant in the fractions smaller than 125 μm. Radionuclides, once deposited on the ground, can be re-entrained into the atmosphere by the wind action and/or by anthropogenic disturbances (e.g. agricultural practice and road traffic).[20] Particularly, considering the ground condi-

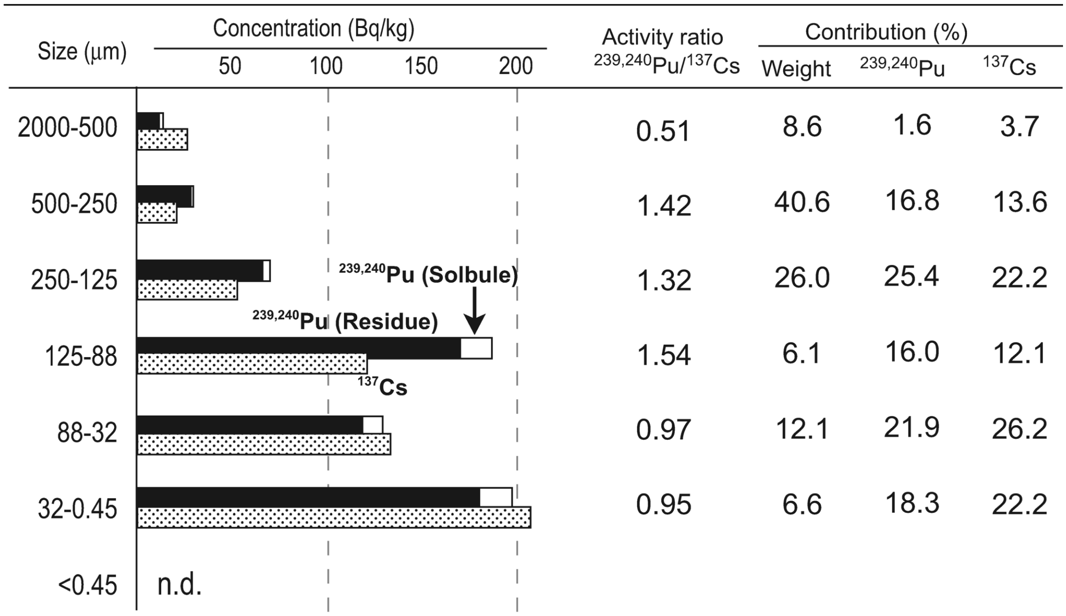

Fig. 3. Particle size dependencies of ^{239,240}Pu and ^{137}Cs in surface soil from Dolon.

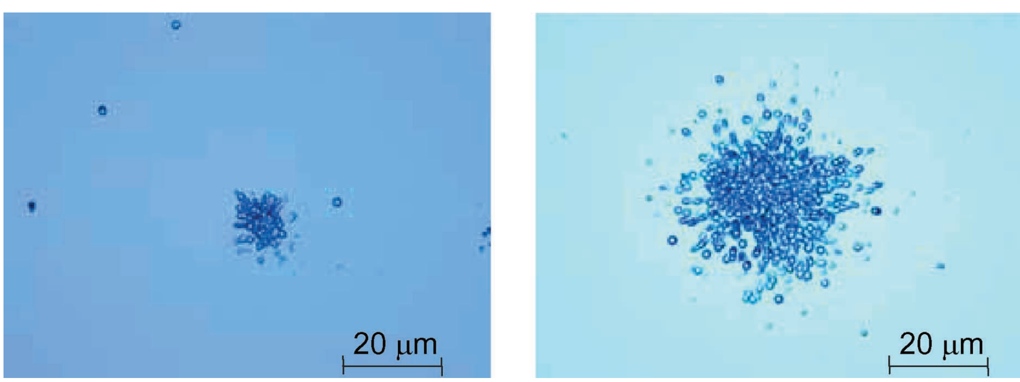

Fig. 4. Alpha-track clusters in surface soil from Dolon (0–10 cm in depth). The sample is No. 24 shown in Fig. 2.

tion such as dryness, sandiness and high wind speed in this area, the findings obtained here will be helpful for evaluating the current and future radiation risks to the people living around here.

Finally it is to note that the data obtained in this study were used recently as a basis for estimating of width and center-axis location of the radioactive plume, in relation to the efforts to reconstruct the radiation dose in Dolon, which was affected by the first USSR atomic bomb test in 1949 at the SNTS.[25)]

CONCLUSION

The current radiological situation at Dolon, Mostik, Cheremushka and Budene settlements near the SNTS territory was investigated using the soil core samples up to a depth of 30 and 100 cm. The results are summarized as follows:

(1) The ^{137}Cs and ^{239,240}Pu inventories varied largely even in a small area, with wide ranges from 140-10,310 Bq/m^2 and 140-14,320 Bq/m^2, respectively.
(2) From the depth distributions of ^{137}Cs and ^{239,240}Pu concentrations, the areas within and close to these villages were rather disturbed by not only human and/or animal activities but also natural phenomenon.
(3) Most of the Pu in soil was tightly incorporated into various sizes of fused particles formed in the course of the condensation of melting materials such as vaporized soil and bomb materials.
(4) Both ^{137}Cs and ^{239,240}Pu in soil were accumulated in the smaller soil size fraction of <125 µm, and presence of hot particles, probably due to Pu, was clearly observed by star-like patterns from α-tracks.
(5) The obtained data will serve as a basis for evaluating the current and future radiation risks to the people living

around here.

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REFERENCES

1. Takada, J., Hoshi, M., Rosenson, R. I., Endo, S., Yamamoto, M., Nagatomo, T., Imanaka, T., Gusev, B. I., Apsalikov, K. N., Tchaijunusova, N. J. (1997) Environmental radiation dose in Semipalatinsk nuclear test site. Health Phys. **73**: 524–527.
2. Gusev, B. I., Abylkassimova, Z. N., Apsalikov, K. N. (1997) The Semipalatinsk nuclear test site: a first assessment of the radiological situation and the test-related radiation doses in the surrounding territories. Radiat. Environ. Biophys. **36**: 201–204.
3. Deliglasow, V. I., Gorin, V. V., Maltzev, A. L., Matusehnko, A. M., Safonov, F. F., Smagulov, S. G. (1991) Radiological situation at Semipalatinsk test site bordering regions of the Kazakh SSR. Bulletin of the public information center by atomic energy (CNII atominform) **N4**: 46–52.
4. Takada, J., Hoshi, M., Nagatomo, T., Yamamoto, M., Endo, S., Takatsuji, T., Yoshikawa, I., Gusev, B. I., Sekerbaev, A. K., Tchaijunusova, N. J. (1999) External doses of residents near Semipalatinsk nuclear test site. J. Radiat. Res. **40**: 337–344.
5. Ivannikov, A. I., Zhumadilov, Z., Gusev, B. I., Miyazawa, Ch., Jiao, L., Skvortsov, V. G., Stepanenko, F. V., Takada, J,. Hoshi, M. (2002) Individual dose reconstruction among residents living in the vicinity of the Semipalatinsk nuclear test site using EPR spectroscopy of tooth enamel. Health Phys. **83**: 183–196.
6. Yamamoto, M., Hoshi, M., Takada, J., Sekerbaev, A. Kh., Gusev, B. I. (1999) Plutonium isotopes and ^{137}Cs in the surrounding areas of the former Soviet Union's Semipalatinsk nuclear test site. J. Radioanal. Nucl. Chem. **242**: 63–74.
7. Sakaguchi, A., Yamamoto, M., Hoshi, M., Apsalikov, K. N., Gusev, B. I. (2004) Plutonium isotopes and ^{137}Cs in Dolon settlement near the Semipalatinsk nuclear test site-about 50 years after the first nuclear weapon testing. J. Radioanal. Nucl. Chem. **260**: 543–555.
8. Gastberger, M., Steinhäusler, F., Gerzabek, M. H., Hubmer, A., Lettner, H. (2000) ^{90}Sr and ^{137}Cs in environmental samples from Dolon near the Semipalatinsk nuclear test site. Health Phys. **79**: 257–265.
9. Gastberger, M., Hubmer, A., Steinhäusler, F., Lettner, H., Spano, H., Stronati, L., Testa, A. (2001) Plutonium in soil from Dolon near the Semipalatinsk nuclear test site. Radiochim. Acta **89**: 371–375.
10. Shebell, P., Hutter, A. R. (1998) Environmental radiation and radioactivity in the vicinity of the Semipalatinsk test site. J. Radioanal. Nucl. Chem. **235**: 133–138.
11. Steinhausler, F., Gastberger, M., Hubmer, A., Spano, M., Ranaldi, R., Stronati, L., Testa, A. (2000) Assessment of the radiation dose due to nuclear tests for residents in areas adjacent to the Semipalatinsk test site "Polygon" (Kazakhstan). Proceedings of IRPA 10 Hiroshima May 2000. P-4b-222.
12. Yamamoto, M., Tsukatani, T., Katayama, Y. (1996) Residual radioactivity in the soil of the Semipalatinsk nuclear test site in former USSR. Health Phys. **71**: 142–148.
13. Yamamoto, M., Tsumura, A., Tsukatani, T. (1998) Current levels of Pu isotopes and ^{137}Cs at the former Soviet Union's Semipalatinsk nuclear test site. Radiochim. Acta **81**: 21–28.
14. Yamamoto, M., Hoshi, M., Takada, J., Oikawa, S., Yoshikawa, I., Takatsuji, T., Sekerbaev, A. Kh., Gusev, B. I. (2001) Some aspects of plutonium in and around the former Soviet Union's Semipalatinsk nuclear test site. Plutonium in the Environment (Kudo, A. Ed.) Elsevier Sciences Ltd. p. 375–400.
15. Yamamoto, M., Hoshi, M., Takada, J., Oikawa, S., Yoshikawa, I., Takatsuji, T., Sekerbaev, A. Kh., Gusev, B. I. (2002) Some aspects of environmental radioactivity around the former Soviet Union's Semipalatinsk nuclear test site: local fallout Pu in Ust'-Kamenogorsk district. J. Radioanal. Nucl. Chem. **252**: 373–394.
16. UNSCEAR (2000) Report of the United Nation Scientific Committee on the effects of atomic radiation, New York.
17. Shapiro, C. S., Kiselev, V. I., Zaitev, E. V. (1998) Nuclear tests, long-term consequences in the Semipalatinsk/Altai region. NATO ASI series, 2. Environment – Vol.36, Springer, Berlin, Heidelberg, Germany.
18. Yamamoto, M., Komura, K., Sakanoue, M. (1983) ^{241}Am and plutonium in Japanese rice-field surface soils. J. Radiat. Res. **24**: 237–249.
19. Kazachevskiy, I. V., Solodukhin, V. P., Chumikov, G. N., Lukashenko, S. N. (1998) Some aspects of determination of radionuclides at the former Semipalatinsk nuclear test site. J. Radioanal. Nucl. Chem. **235**: 145–149.
20. Sehmel, G. A. (1973) Particle resuspension from an asphalt road caused by car and truck traffic. Atmosph. Environ. **7**: 291–301.
21. Hille, R., Hill, P., Bouisset, P., Calmet, D., Kluson, J. (1998) Population dose near the Semipalatinsk test site. Radiat. Environ. Biopys. **37**: 143–149.
22. Duvasov, Y., Krivohatskii, A., Kharitonov, V., Gorin, V. (1994) Radioactive contamination of the Semipalatinsk testing ground and adjacent territories in consequence of atmospheric nuclear tests in 1949–1962. International Symposium on Remediation and Restoration of Radioactive - Contaminated Site in Europe, 11–15 Oct. 1993, Proceedings, Antwerp.
23. Deriglasov, W. N., Safonov, F. F., Smagulov S. O., Jurtschenko, T. I. (1993) Radiohygenic Situation in the Region of the Semipalatinsk Polygon. Bulletin of the Centre of Public Information on Nuclear Energy, Moscow. Special issue: 20 January 1993.
24. Voigt, G. M., Semiochkina, N., Dodd, B., Howard, B. J., Karabalin, B., Mukuschewa, M., Rosner, G., Sanchez, A., Singleton, D. L., Strand, P. (2001) The present radiological situation at the nuclear weapons test site at Semipalatinsk in

Kazakhstan with regard to plutonium contamination. Plutonium in the Environment (Kudo, A. Ed.) Elsevier Sciences Ltd. p. 363–373.

25. Imanaka, T., Fukutani, S., Yamamoto, M., Sakaguchi, A., Hoshi, M. (2005) Width and center-axis location of the radioactive plume that passed over Dolon and nearby villages on the occasion of the first USSR A-bomb test in 1949. J. Radiat. Res. **46**: 395–399.

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