Dietary intakes and internal exposure doses received by residents of the Karachai Trace

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Abstract. Due to several radioactive accidents at the Mayak Production Association, a facility for production of weapon plutonium put on line in 1948 in the Southern Urals (Russia), an substantial part of the Southern Urals territory was contaminated by long-lived radionuclides. One of the such accident was the wind resuspension of radionuclides ($2{\cdot}10^{14}$Bq) from the shoreline of Karachai lake used as a depot of liquid radioactive waste, that formed the Karachai Trace in 1967. The main task of the present investigation is to estimate the dietary intakes of radionuclides and exposure doses for residents of settlements located on the Karachai Trace. For these purposes, dynamics of changes in the contents of ^{90}Sr and ^{137}Cs in main foodstuffs and diet composition were studied for residents of 6 settlements. Milk was found to be the main contributor of radionuclides to the residents' diet. The pattern of the decrease in radionuclide concentration in milk can be described by a two-exponential dependence. The biological accessibility of ^{90}Sr and ^{137}Cs settled in the Karachai area was lower compared to that observed on the East-Urals Radiation Trace (formed in 1957 due an explosion in the radioactive waste-storage facility), or in the Chernobyl area. Accumulated and current exposure doses were estimated for each calendar year based on the data on contents of radionuclides in the residents' diets. The accumulated whole body dose from ^{137}Cs estimated for adult residents in the most heavily contaminated village of Sarykulmiak was 0.83 mSv. The accumulated red bone marrow dose from ^{90}Sr was 19.1 mSv. The summary effective dose from ^{137}Cs and ^{90}Sr amounted to 3.7 mSv.

1. Introduction

The Mayak production association, created in 1948 for production of plutonium for nuclear weapons, was the source of radionuclide environment contamination in the Southern Urals (Russia). Most important of radiation accidents were the releases of radioactive material (10^{17} Bq) into the Techa River (1949-1956); an explosion in the radioactive waste-storage facility in 1957, that formed the East Urals Radioactive Trace (EURT) due to dispersion $7.4{\cdot}10^{16}$ Bq into the atmosphere; and wind transfer of radionuclides ($2.2{\cdot}10^{14}$ Bq) from the waste settling reservoir (Karachai Lake) in 1967, that formed the Karachai Trace. Karachai trace, which is a focus of present investigation, was formed in April 1967 as a result of a downwind transfer of shoreline and bottom sediment from the partly dried out Karachai Lake.

The wide range of environmental contamination levels in the Urals Region provides an opportunity to investigate the specific features of environmental transfer of radionuclides depending on the soil type, physical and chemical form of radionuclides, contamination density of soil, etc. The collection and measurements of environmental samples and local foodstuffs in the contaminated areas of the Urals Region were started by the researchers of the Urals Research Centre for Radiation in 1958. The results of the measurements constitute the basis for the Data Base "Environment" which was created in 1998 to allow summarization as a result of environmental monitoring conducted of information collected in the period from 1958 to 2003. The Data Base includes the information on the contamination of the environment, soil, river water and food on the East Urals Radioactive Trace, Karachai Trace, as well as data on the global fallout. Currently, the Data Base allows making analysis of the relationship between soil, grass and foodstuff contamination at different localities in the Southern Urals. As for the impacts of the Karachai Trace, such analysis can be performed for the period over the 30 years.

The main purpose of the paper is to identify the role of Karachai Trace in the radioactive contamination of the Southern Urals and to estimate its contribution in the public exposure. Thus, the primary tasks are the following: (1) summarize the data on ^{90}Sr and ^{137}Cs diet contamination for the residents of the Karachai Trace over the total period since the radiation accident; (2) verify the reconstructed dietary intake function; (3) estimate internal doses of public exposure due to ^{90}Sr and ^{137}Cs.

1. Materials and methods

The isotope composition of the settled particles in Karachai Trace was as follows: $^{90}Sr+^{90}Y$ - 32 %, ^{137}Cs - 47 %, $^{144}Ce+^{144}Pr$ – 21 % [1] which differs significantly from the composition of the fallout due to EURT. It was established based on the studies of the fractional composition of the dust gathered from the vegetation that 10-50 μn accounted for 50 % of the mass, 1-10 μn for 11,2 %, and 1,5 % are constituted of <1 μn fraction made up of silt. Biological accessibility of ^{90}Sr was higher than that for ^{137}Cs by a factor of 10. The area of the trace bounded by the isoline with contamination density for ^{90}Sr above 3.7 $kBq{\cdot}m^{-2}$ amounted to 1800 km^{-2}. Radionuclides settled on this territory to totaled about $2.2{\cdot}10^{14}$ Bq.

The source of radioactive contamination for both East Urals Radioactive Trace and Karachai Trace was Mayak production association, and the territory of the traces are partly overlap one another. So, for sample collection 6 settlements located on the territory predominantly contaminated due to Karachai Trace were selected. The 6 settlements are located in the different distance from the source of contamination (Table I).

Table I. Characterization of the villages where the sampling was performed.

Names of villages	Distance from the source of contamination, km	Number of population as of 1970	Contamination density for ^{90}Sr, $kBq{\cdot}m^{-2}$, 1967	Contamination density for ^{137}Cs, $kBq{\cdot}m^{-2}$, 1967	Period of sampling
Kainkul	35	320	32.1	96.2	1967-2000
Karagaikul	27	627	20.9	62.9	1967-2001
Kunashak	45	4813	16.4	37	1967-1977
Sary	35	1203	17.2	51.8	1967-1985
Sultayevo	40	737	12.3	37.0	1967-1972
Sarykulmiak	18	606	54.8	161.8	1967-2001

In these settlements the basic foodstuffs (milk, meat, potato) were produced at their private farms. First samples of foodstuffs began to be taken from these farms soon after the radionuclides settled on the soil. Samples of potato were collected during the harvest time. The total samples collected over the entire period included 300 specimens of soil from kitchen-gardens and pasturing lands, 1,550 samples of milk, and 270 samples of potato. In the period from 1967 through 1977 samples of milk were taken during the pasturing and stabling period and since 1985 during the pasturing period only. The collection of milk samples for early years (1967-1977) were performed each month of the year, for subsequent years the samples were taken in summer season.

The food samples were ashed and measured using radiochemical method for ^{90}Sr and ^{137}Cs [2]. The concentration of ^{137}Cs was also determined by gamma-spectrometry [3].

Radionuclide body contents in human were measured using the whole-body counters (WBC) SICH–9.1 (URCRM, Chelyabinsk in 1977) and SICH–2.2 (Moscow, Institute of Biophysics) in 1969.

Internal doses due to ^{137}Cs were calculated in accordance with ICRP Publication 67 [4]. The calculation of doses of ^{90}Sr was made using the DosAge software, developed at the URCRM in 1992 on the basis of an age-specific biokinetic model for ^{90}Sr [5]. Mathematical processing of data was carried on using the Excel 97 and Origin 50 software.

3. Results

3.1. Reconstruction of radionuclide intakes

3.1.1. Radionuclide contamination of milk

In order to use the most appropriate mathematical model for averaging of the results of radionuclide measurements, the statistical distributions of ^{90}Sr and ^{137}Cs concentration in milk samples were analyzed. Fig. 1 and Fig. 2 exemplified statistical distribution of ^{90}Sr and ^{137}Cs contents in milk samples. It was shown that radionuclide concentration in milk samples were described by log-normal distribution with sufficient accuracy. So, the geometrical mean values of radionuclide measurements in foodstuffs were used for estimating average dietary intakes.

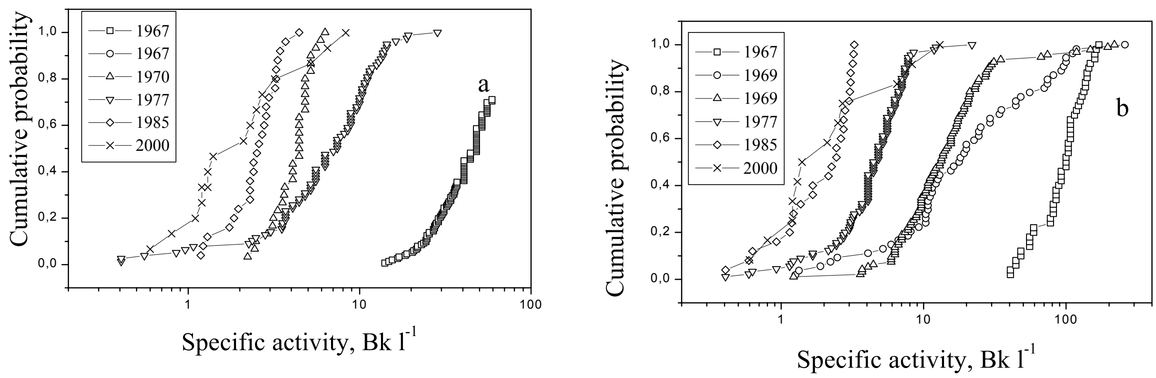

FIG. 1. Probabilistic-statistical distribution of specific activity of ^{90}Sr (a) and ^{137}Cs (b) in milk samples from the village of Sarykulmiak.

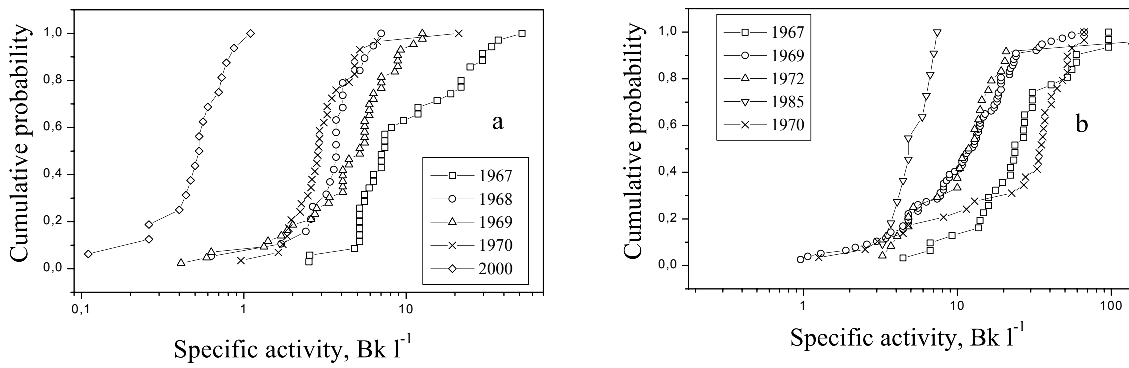

FIG 2. Probabilistic-statistical distribution of specific activity of ^{90}Sr (a) and ^{137}Cs (b) in milk samples from the village of Kainkyl.

Fig. 3 shows the dynamics of radionuclide contents in milk (mean levels for the pasturing period) produced in the village of Sarykulmiak. As cab be seen, the initial specific activity of ^{90}Sr in milk measured late in April 1967 made up 140 $Bq \cdot l^{-1}$, that of ^{137}Cs was 237 $Bq \cdot l^{-1}$. From 1967 through 2001 mean values of specific activities of ^{90}Sr and ^{137}Cs in milk decreased 30-fold, on the average. The decreases with time in contents of ^{90}Sr and ^{137}Cs in milk can be described by 2 exponents. The period of half-purification ($T_{1/2)}$) of milk during the early period (the first exponent) was influenced by reductions in radioactive contamination of grass surface and it lasted 0.3-0.5 years. The second exponent describes a decreased content of radionuclide in milk due to a decreased amount contributed by soil. The process of vertical migration in the soil and changes of biological accessibility of radionuclides influence to this process. The periods of half-purification ($T_{1/2\ eff.}$) were 20 years for ^{90}Sr, and 10 years for ^{137}Cs.

The time period needed for milk to be purified of ^{137}Cs on the Karachai Trace is comparable to that observed in the area contaminated due to the Chernobyl accident [6]. The time needed for milk to be purified of ^{90}Sr on the Karachai Trace is close to the value of the radionuclide content reduction in milk observed in the EURT area [7].

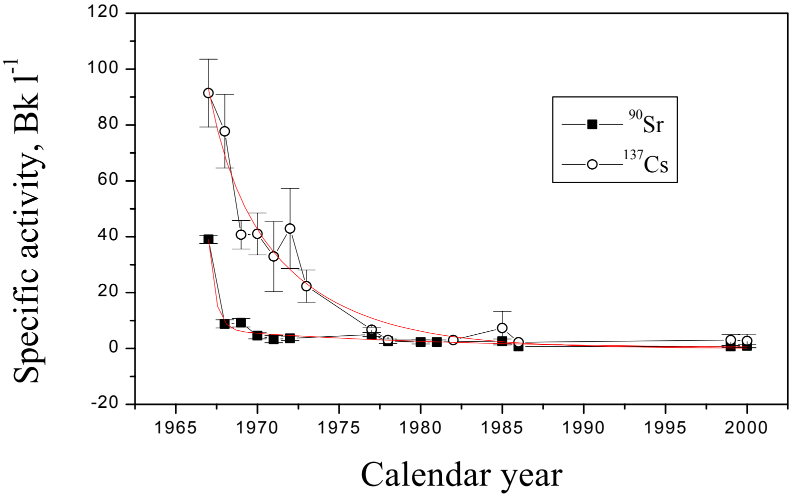

FIG. 3. Changes in specific activity of radionuclides in milk sampled in the village of Sarykulmiak. Vertical lines limit the mean value error.

3.1.2. Transfer coefficient from soil to milk

The migration of radionuclides along the soil-milk chain is characterized by transfer coefficients (TC) defined as the relationship between mean contents of ^{90}Sr and ^{137}Cs in milk and the mean content of these radionuclides in the soil of the pastures the cattle grazed on (Fig. 4).

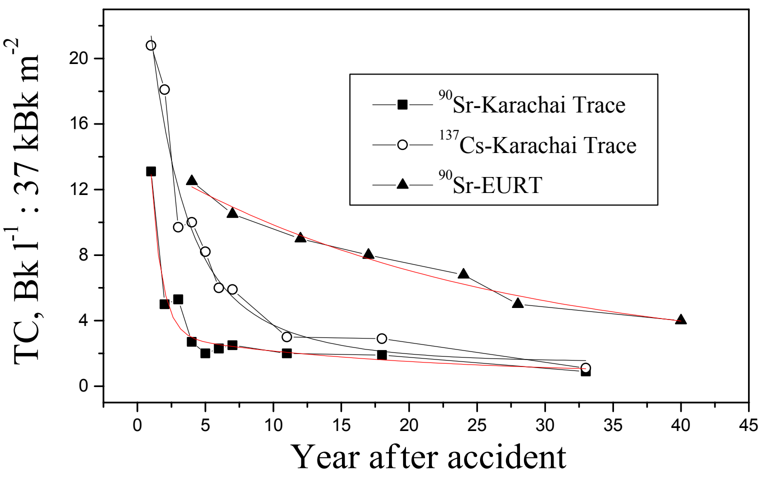

FIG. 4. Changes in transfer coefficients in the soil-milk system of the Karachai Trace and EURT as a function of time period since the fallout of the radionuclides.

The values of TC for ^{137}Cs in the soil-milk system estimated for the first post-accident year on the Karachai Trace was 5 times lower than that estimated for a number of regions contaminated due to the

Chernobyl accident [6] for which the TC values fluctuated from 55 to 85 $Bq \cdot l^{-1}$ per 37 $kBq \cdot m^{-2}$. In the ensuing years the TC value estimated for ^{137}Cs in the soil-milk chain on the Karachai Trace was also much lower compared to that observed on the territory contaminated due to the Chernobyl accident [8].

Transfer coefficients estimated for ^{90}Sr in the soil-milk chain on the Karachai Trace was, on the average, 5-fold lower compared to those on the EURT determined for the respective post-accident time intervals. A reduced migration of ^{90}Sr and ^{137}Cs from forage to cow milk during the first pasturing period on the Karachai Trace was noted in the publication [9] too. Evidently, the long time period that ^{90}Sr and ^{137}Cs remained deposited in the soils and bottom sediments of the Karachai Lake shoreline resulted in a reduced biological accessibility of radionuclides for plants.

3.1.3. Radionuclide contamination of potatoes

Table II exemplifies the average concentration of ^{90}Sr and ^{137}Cs in potatoes in Sarykulmiak, which is placed in the most contaminated territory of the Karachai Trace.

Table II. Specific activity of radionuclides in samples of potatoes *from the village of Sarykulmiak, $Bq \cdot kg^{-1}$*

Year	^{90}Sr	^{137}Cs
1967	1±0.4 (3)	17±10.6 (6)
1968	1.5±1.8 (8)	4.7±7.2 (8)
1969	1.5±1 (9)	2.8±1.2 (8)
1970	0.8±1.1 (30)	2±1.4 (24)
1971	0.9±0.8 (25)	1.7±0.9 (25)
1980	0.8±0.6 (30)	0.8±0.6 (30)
1985	0.8±0.5 (19)	1.5±0.8 (21)
1999	0.6±0.1 (19)	1±0.8 (15)

Note: - In parenthesis: the number of measurements

As can be seen, in Sarykulmiak over the total period the specific activity of ^{90}Sr contained in vegetables and potato decreased 2-fold, and that of ^{137}Cs - 17-fold. The mean values of specific activities of ^{90}Sr contained in potato in others villages were found to be low and did not exceed 1.5 $Bq \cdot kg^{-1}$ during the all period of observation. The radionuclide contents in samples of potato from different villages differed up to 3-fold. Cesium-137 contents in potatoes from other villages were also lower than in Sarykulmiak and did not exceed 5 $Bq \cdot kg^{-1}$.

3.1.4. Radionuclide contamination of meat and bread.

Specific activities of radionuclides in samples of meat for the period 1967-1970 were measured in a limited number of tests; the results are summarized in Table III.

Table III. Specific activity of radionuclides measured in meat samples (x±m), $Bq \cdot kg^{-1}$

Sampling site	1967-1968	1968-1969	1969-1970
	^{90}Sr		
v. Sarykulmiak	2±1.7 (4)	9±11 (8)	-
Others villages	2±1 (8)	-	-
	^{137}Cs		
v. Sarykulmiak	24.8±10 (4)	21.5±13 (8)	41±37 (5)
Others villages	0.6±0.3 (6)	-	-

Note: - In parenthesis: the number of measurements

The radionuclide concentrations in milk and beef in the same period of time were rather similar (Fig.3). So, to allow assessment of average dietary intakes of radionuclides with meat it was assumed that the concentration of ^{90}Sr and ^{137}Cs in meat and milk were the same.

The radionuclide content in bread was mainly attributed to global contamination and was not taken into account in the radionuclide intake reconstruction.

3.1.5. Average radionuclide dietary intake in the period 1967-2002

The average composition of daily diet for Southern Urals villages is presented in Table IV.

Table IV. Diet composition for adult village residents in Southern Urals.

Food-staff	**Consumption, gram per day**
Milk and milk products	620
Bred and grain products	600
Meat products	140
Potato	276
Vegetables	167

Based on these data and data on local foodstuff contamination the annual radionuclide intakes were calculated for the period from 1967 to 2001 (Fig. 5).

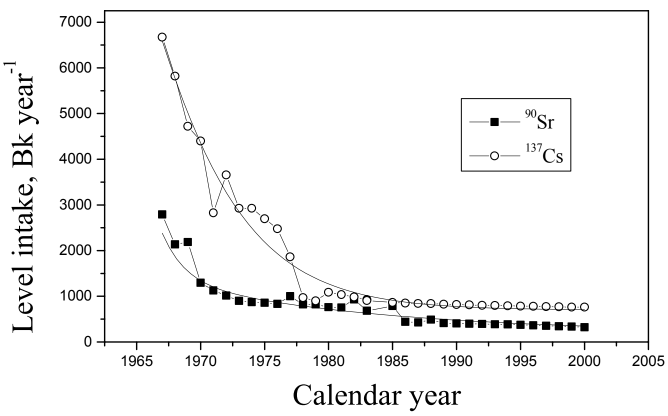

FIG. 5. The annual levels of intake of radionuclides for adult residents of Sarykulmiak.

As can be seen, during the period from 1967 through 1977 the dietary intake of ^{137}Cs in adults was 2-3 times higher than that of ^{90}Sr. In later years the difference in annual intakes of the radionuclides was no more than 1.5 times. The difference in the contents of radionuclides in diet observed in the early and late periods attributed to the difference in transfer coefficients from soil to milk and soil-vegetable for ^{137}Cs and ^{90}Sr.

3.1.6. Verification of radionuclide intake functions

The human measurements of ^{137}Cs body contents allows to verify the intake function for ^{137}Cs. For this purpose, mean-annual ^{137}Cs body intakes (Fig. 5) were incorporated into the ICRP-67 model which was used to estimate ^{137}Cs body contents for individuals in different age categories. Calculated values were compared with the results of measurements (Table V).

Table V. Comparison of ^{137}Cs body contents for Sarykulmiak residents estimated on the basis WBC measurements and model calculation

Method of estimation of cesium-137 body content	**Cesium-137 body content, Bq**	
	Children aged 4-14 years, 1969	**Adults, 1977**
WBC measurements	833 (322-1344)[a]	555 (296-814) [a]
Model calculation based on reconstructed ^{137}Cs intakes (Fig. 5)	977 (425-3059) [b]	580 (250-1335)[c]

[a] - In parenthesis: the range of standard deviation, [b] - In parenthesis the range of calculated ^{137}Cs body contents for peoples of different ages taken into account the uncertainties of ^{137}Cs intake reconstruction (minimal and maximal values),
[c] - In parenthesis the range of ^{137}Cs body contents calculated taken into account the uncertainties of ^{137}Cs intake reconstruction (minimal and maximal values)

As can be seen in Table V, the estimated radionuclide body contents are comparable with the measured ones and can be used for internal dose calculation. On the average, the ^{137}Cs body contents measured in 1969 for children were 6 times higher than those determined for adult rural residents of the USSR [10]. The corresponding ratio for adults in 1977 was 4.4.

3.2. Doses of radioactive exposure in the territory of Karachai Trace

Since the village of Sarykulmiak was investigated more completely, it was designated as the reference village for the exposed population. Estimated ^{137}Cs dose accumulated by adult Sarykulmiak residents over the period 1967-2001 was 0.83 mSv. As for bone-seeking ^{90}Sr the read bone marrow doses are most important in terms of biological and health effects. The cumulative absorbed red bone marrow dose from ^{90}Sr was 19.1 mSv for adults. The cumulative effective dose from ^{137}Cs and ^{90}Sr summary was estimated to be 3.7 mSv, and the current annual effective dose (2001) was estimated as 0.03 mSv.

To allow exposure dose estimation for residents of other villages doses were calculated per unity contamination density. For this purpose the doses calculated for Sarykulmiak residents were referred to the contamination density registered in the locality. The contamination density in the Sarykulmiak pasturing area made up 54.8 $kBq\cdot m^{-2}$ for ^{90}Sr and 162 $kBq\cdot m^{-2}$ for ^{137}Cs. The cumulative red bone marrow dose contributed by ^{90}Sr was 0.35 mSv per 1 $kBq\cdot m^{-2}$. The cumulative ^{137}Cs whole-body dose was 0.005 mSv per 1 $kBq\cdot m^{-2}$ of ^{137}Cs.

Taking into account the initial contamination density (Table I) and the obtained referent values, the average accumulated doses from ^{90}Sr and ^{137}Cs were calculated for other villages: Sultayevo, Sary, Kainkul, Karagaikul and Kunashak. The exposure doses received by the residents were lower by a factor of 3 than those for Sarykulmiak.

4. Discussion

The two accidents that occurred at the Mayak facility resulted in contamination of significant agriculture lands in Chelyabinsk oblast: the 1957 explosion and wind transport of radionuclides from Karachai Lake in 1967. A detailed monitoring was performed for both radioactive traces, which partially superimpose one another. The contamination due to explosion of 1957 was significantly higher than that of 1967. So, it was difficult to separate the role played by the two accidents in the contamination of specific settlements. It should be taken into account that the physical-chemical form of radionuclides differed significantly in these accidents, but strontium-90 was one of main source of exposure in both situations.

The study of ^{90}Sr transfer from soil to grass, vegetables and then to cow milk/meat in the contaminated area allows comparisons of intake functions for EURT and Karachai Traces depending on the time after the accidents. Fig. 6 shows the normalized intake function for EURT and Karachai territory in terms of annual radionuclide intakes per unit of ^{90}Sr contamination density. As can be seen, for early period after contamination the difference between intake functions was about one order of magnitude. It is important for persons who lived on the territories where there was a superposition of the two radioactive traces. So, the relative contribution of doses from additional "Karachai" contamination to the total doses of human exposure can not be significant for such residents.

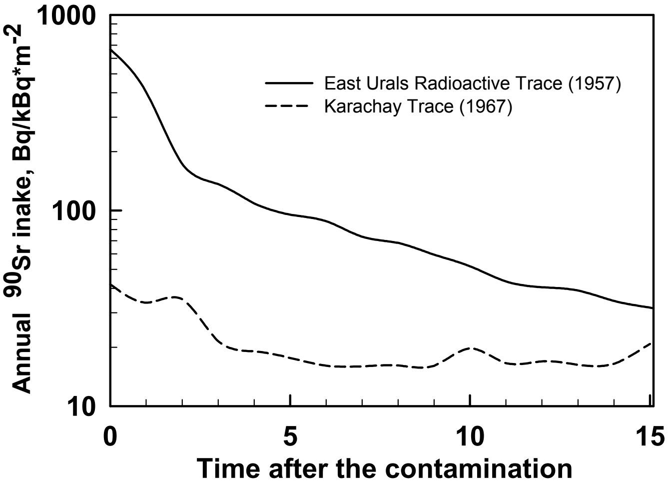

FIG. 6. The normalized annual ^{90}Sr intakes for adult residents EURT and Karachai territories.

5. Conclusion

The long time period that the radionuclides ^{90}Sr and ^{137}Cs had been incorporated in the composition of soil and silt of the shores of the Karachai Lake resulted in their transformation into physical-chemical states less accessible to plants, and consequently, in reduced dietary intakes with milk and other foodstuffs as compared with the intakes per unit of contamination density due to the East-Urals Trace and Chernobyl accident.

The cumulative effective dose from ^{137}Cs and ^{90}Sr estimated for adult residents of Sarykulmiak over the 33 years since the Trace was formed is 3.7 mSv, and that for residents of Sultayevo, Sary, Kainkul, Karagaikul and Kunashak is 1.3 mSv.

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