Environmental Radioactivity in Hungary

Bulletin No. 2

EDITED BY

J. KOVÁCS AND

T. PREDMERSZKY

BUDAPEST 1979

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PREFACE

The information on data of environmental radioactivity available in Hungary is proceeding with the spirit of Bulletin No.1. The publication aims at rendering comprehensive results on measuring data reported in special reviews, conferences and other forums. If a complex serie of measurement is available all data of the full serie are presented. If not so, we report on those that were not mentioned in Bulletin No.1. At present, in addition to data of artificial radionuclides, we report on environmental data with respect to natural radioactivity. Considering that the data were given in Bulletin No.1 in former units of measurement, the recalculation into SI-units has been neglected this time.

J. Kovács - T. Predmerszky

Investigation of Environmental Radioactive Contamination in Hungary

1. STUDIES ON ATMOSPHERIC RADIOACTIVE CONTAMINATION

Fall-out Measurements

The examination of the radioactive contamination of atmospheric fall-out, i.e. the gross beta activity, has been regularly carried out in Debrecen since March, 1952. The results of the measurements between 1952 and 1976 are shown in Fig. 1. /1/.
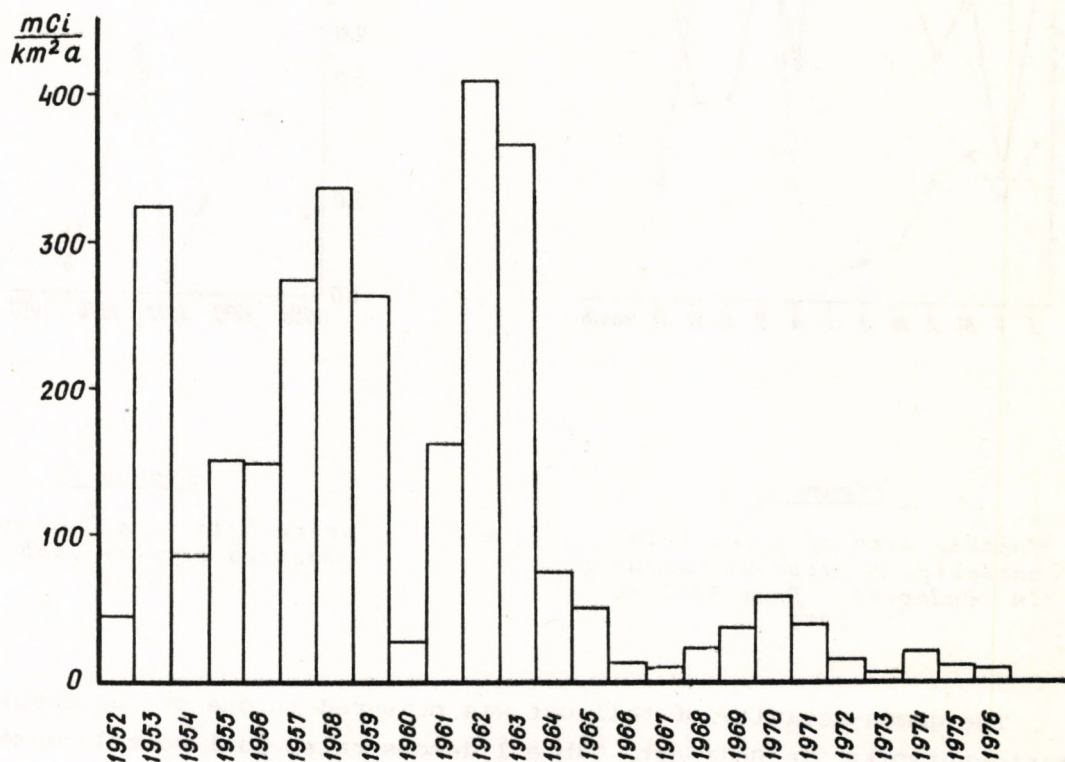
Figure 1

Beta activity of fission products in the atmospheric precipitation in Debrecen, Hungary, during 1952 and 1976

Monthly data of gross beta activity of fall-out measured in Budapest from 1975 to 1976 are shown in Fig. 2 [2].
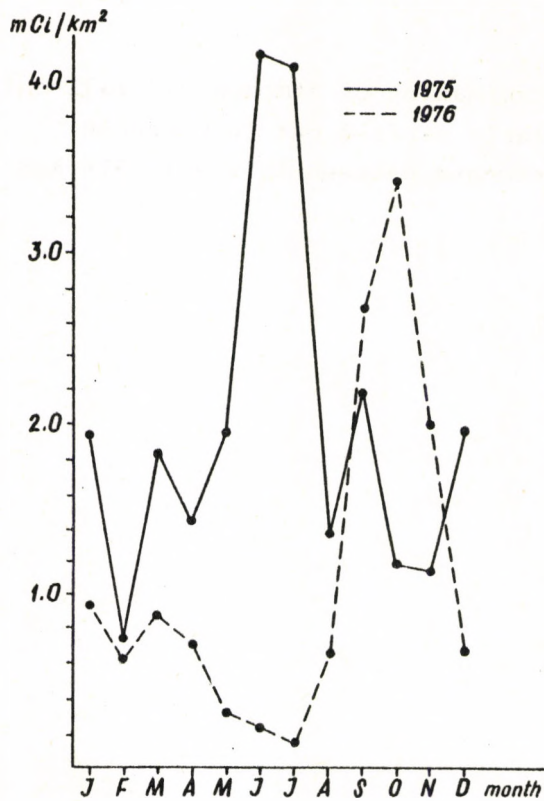
Figure 2

Monthly data of gross beta activity of fall-out measured in Budapest from 1975 to 1976
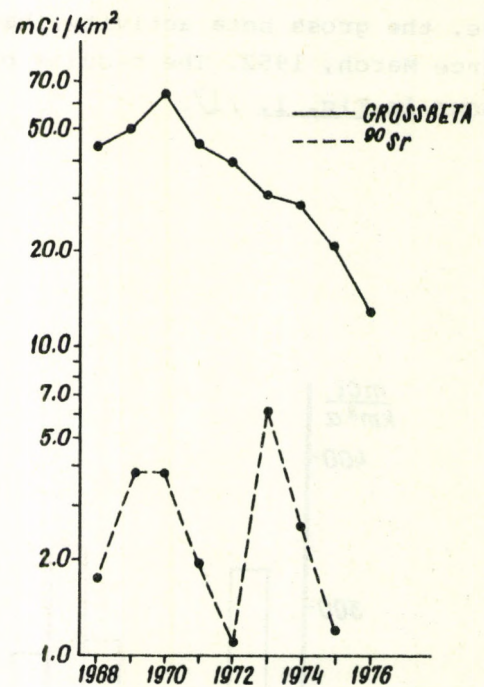
Figure 3

Gross beta- and ^{90}Sr activity measured between 1968 and 1976

The gamma-activity of fall-out was measured in one of the sampling stationnetworks in Budapest. Table 1 demonstrates full monthly data measured at this station between 1964 and 1976 [3].

Year / Month	1964	1965	1966	1967	1968	1969	1970	1971	1972	1973	1974	1975	1976
January	–	0.9	0.4	0.5	0.4	0.3	0.6	0.2	0.5	0.5	1.1	0.3	<0.2
February	–	0.3	0.5	0.8	<0.1	0.2	0.3	0.3	0.3	<0.2	0.3	0.5	<0.2
March	–	0.3	0.2	0.5	0.4	0.2	< 0.2	1.1	0.2	0.3	0.5	0.8	<0.2
April	–	0.9	0.1	0.3	0.4	0.3	0.5	1.1	2.7	0.3	0.5	0.4	<0.2
May	–	0.6	0.5	< 0.1	0.7	0.5	1.5	0.8	2.6	0.2	1.1	0.8	<0.2
June	13.3	2.7	0.3	0.2	0.5	0.5	1.3	2.1	1.7	0.4	0.7	0.4	0.2
July	6.5	1.5	0.9	0.2	0.4	1.4	3.2	1.3	1.0	0.3	0.4	< 0.2	<0.2
August	2.7	1.1	0.2	0.1	0.5	0.5	1.9	1.4	0.4	<0.2	0.4	< 0.2	<0.2
September	1.5	0.5	0.2	0.2	0.6	0.6	0.6	2.4	0.3	<0.2	1.4	< 0.2	0.3
October	4.0	<0.2	0.3	0.2	0.3	0.2	< 0.2	0.4	0.2	<0.2	0.2	< 0.2	4.5
November	1.8	0.3	1.4	0.1	0.3	0.2	< 0.2	1.2	0.2	<0.2	0.3	< 0.2	0.9
December	1.2	< 0.2	0.4	<0.1	0.2	0.4	< 0.2	0.3	0.8	0.5	0.3	< 0.2	0.5

Table 1

Monthy averaged fall-out gamma-activity measured in the region of the Central
Research Institute for Physics between 1964 and 1976

The National Meteorological Office carried out the sampling of the fall-out of precipitation by internationally developed methods of measuring. Tables 2 and 3 demonstrate the gross beta-activity of the fall-out and the precipitate with regard to Budapest, Pécs and Szeged in the years from 1955 to 1976 [4].

Year	Activity, mCi/km^2		
	Budapest	Pécs	Szeged
1955	0.286	–	–
1956	0.521	–	–
1957	0.518	–	–
1958	0.891	–	–
1959	0.600	–	–
1960	0.095	–	–
1961	0.486	–	–
1962	1.845	–	–
1963	3.747	–	–
1964	0.401	0.620	0.410
1965	0.225	0.311	0.187
1966	0.130	0.140	0.040
1967	0.080	0.170	0.060
1968	0.090	0.110	0.050
1969	0.086	0.94	0.035
1970	0.150	0.150	0.060
1971	0.140	0.130	0.070
1972	0.121	0.162	0.046
1973	0.023	0.212	0.039
1974	0.055	0.387	0.050
1975	0.030	0.386	0.153
1976	0.050	0.318	0.112

Table 2

Mean annual values of the gross beta-activity of fall-out in mCi/km^2 [4]

Year	Activity, nCi/1		
	Budapest	Pécs	Szeged
1955	0.16	-	-
1956	0.42	-	-
1957	0.35	-	-
1958	0.65	-	-
1959	0.44	-	-
1960	0.02	-	-
1961	0.21	-	-
1962	1.27	-	-
1963	1,81	-	-
1964	0.20	0.21	0.21
1965	0.08	0.09	0.07
1966	0.03	0.06	0.03
1967	0.04	0.09	0.02
1968	0.05	0.04	0.02
1969	0.04	0.05	0.02
1970	0.08	0.06	0.02
1971	0.12	0.13	0.05
1972	0.08	0.10	0.04
1973	0.02	0.17	0.05
1974	0.03	0.10	0.02
1975	0.02	0.16	0.05
1976	0.02	0.12	0.05

Table 3

Mean annual values of the gross beta-activity of precipitation in nCi/l [4]

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Figure 4 [4] represents the annual mean value of precipitation activity data measured in Budapest as well as the maximum value measured in the current year. It can be observed that the measured maximum values may be higher even by one or two orders of magnitude than the annual mean value.
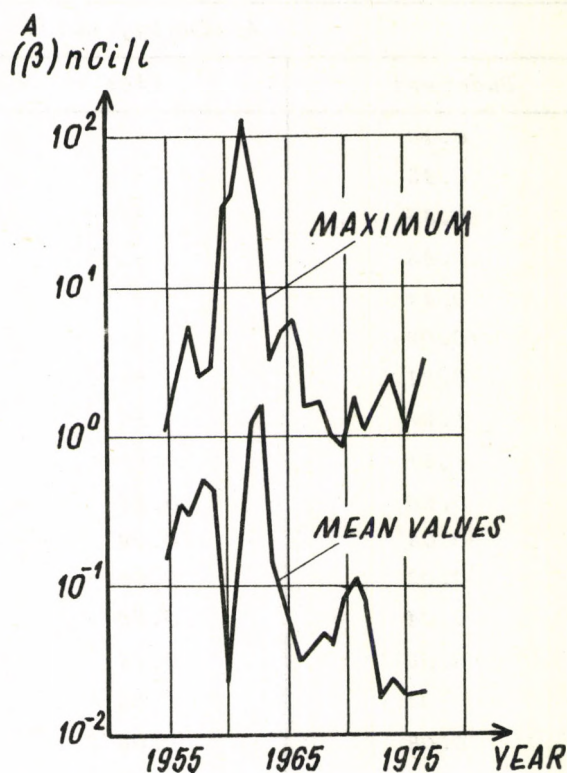
Figure 4

Comparison of the measured maximum radioactivity of precipitation with the annual mean value [4]

Aerosol Measurements

The systematic investigation of the radioactive aerosol contamination close to the surface of the soil took a start within the scope of one of the sampling station networks in Budapest in 1961. The monthly data of the measurements between 1961 and 1976 are shown in the Table 4 [3].

Year / Month	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972	1973	1974	1975	1976
January	–	0.80	3.45	0.40	0.20	<0.10	<0.10	<0.10	0.07	0.08	0.10	0.33	0.10	0.05	0.14	<0.05
February	–	2.03	2.30	0.60	0.10	0.11	<0.10	<0.10	0.12	<0.05	0.09	0.10	0.06	0.10	0.11	0.05
March	–	2.02	1.70	0.50	0.16	<0.10	0.10	0.14	0.13	0.06	0.11	0.09	0.06	0.13	0.15	0.09
April	–	2.30	5.45	1.10	0.16	<0.10	0.10	0.18	0.13	0.09	0.23	0.08	<0.05	0.18	0.14	0.06
May	–	2.70	4.10	1.20	0.20	0.14	0.10	0.23	0.13	0.20	0.30	0.09	<0.05	0.17	0.12	0.05
June	–	2.57	5.07	1.00	0.94	0.31	<0.10	0.22	0.20	0.26	0.27	0.31	<0.05	0.19	0.09	0.06
July	–	2.06	5.45	0.90	0.20	0.10	<0.10	0.09	0.25	0.23	0.23	0.27	0.07	0.16	0.09	0.06
August	–	1.78	2.68	0.50	0.20	0.13	<0.10	0.11	0.24	0.24	0.26	0.21	0.05	0.13	0.07	0.05
September	3.97	3.01	1.67	0.30	0.10	<0.10	<0.10	0.11	0.17	0.16	0.20	0.16	<0.05	0.10	0.09	0.05
October	3.31	3.90	0.90	0.20	<0.10	0.12	<0.10	0.10	0.15	0.14	0.24	0.12	0.05	0.09	0.06	0.52
November	2.75	7.31	0.90	0.30	<0.10	<0.10	<0.10	0.06	0.08	0.11	0.16	0.13	0.05	0.10	0.09	0.08
December	1.10	3.09	0.40	0.10	<0.10	0.10	<0.10	0.06	0.07	0.07	0.12	0.13	0.05	0.07	0.08	0.03

Table 4

Monthly averaged gross beta activity concentration values measured in the atmospheric region of the Central Research Institute for Physics between 1961 and 1976

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Figure 5

*Monthy average of aerosol activity
in pCi/m³ for 1975-1976 [2]*
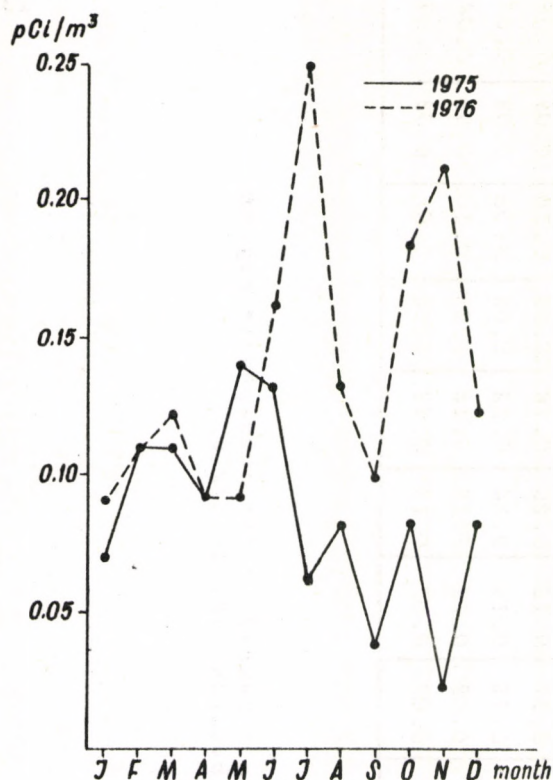
Another station in Budapest presented some changes of the aerosol activity in 1975 and 1976, as seen in Fig. 5 [2].

The determination of the aerosol activity is made in the Central Meteorological Office under internationally agreed circumstances. Table 5 demonstrates those data of aerosol gross beta-activity measured in Budapest, Pécs and Szeged in the years from 1955 to 1976. Maximum and mean values measured in Budapest are shown in Fig. 6.

Figure 6

*Comparison of the maximum aerosol radioactivity with the mean annual
value [4]*

Year	Activity, pCi/m^3		
	Budapest	Pécs	Szeged
1955	1.25	-	-
1956	2.33	-	-
1957	1.81	-	-
1958	3.97	-	-
1959	4.50	-	-
1960	0.30	-	-
1961	1.66	-	-
1962	3.50	-	-
1963	3.29	-	-
1964	0.84	0.66	0.82
1965	0.25	0.27	0.28
1966	0.16	0.18	0.23
1967	0.13	0.07	0.18
1968	0.17	0.11	0.17
1969	0.22	0.17	0.07
1970	0.25	0.10	0.25
1971	0.25	0.09	0.26
1972	0.16	0.05	0.22
1973	0.13	0.04	0.21
1974	0.15	0.10	0.27
1975	0.12	0.10	0.23
1976	0.12	0.17	0.22

Table 5

Annual mean values of the beta-activity of aerosol in
pCi/m³ [4]

Measurement of Radioactive Noble Gas ^{85}Kr Content of the Air

A continuous increase of the ^{85}Kr concentration in the air can be observed all over the world. This fact is of special importance since the operation of fuel reprocessing plants and new establishments make a considerable influence on ^{85}Kr concentration. ^{85}Kr activity measurements are shown in Fig. 7 [5].
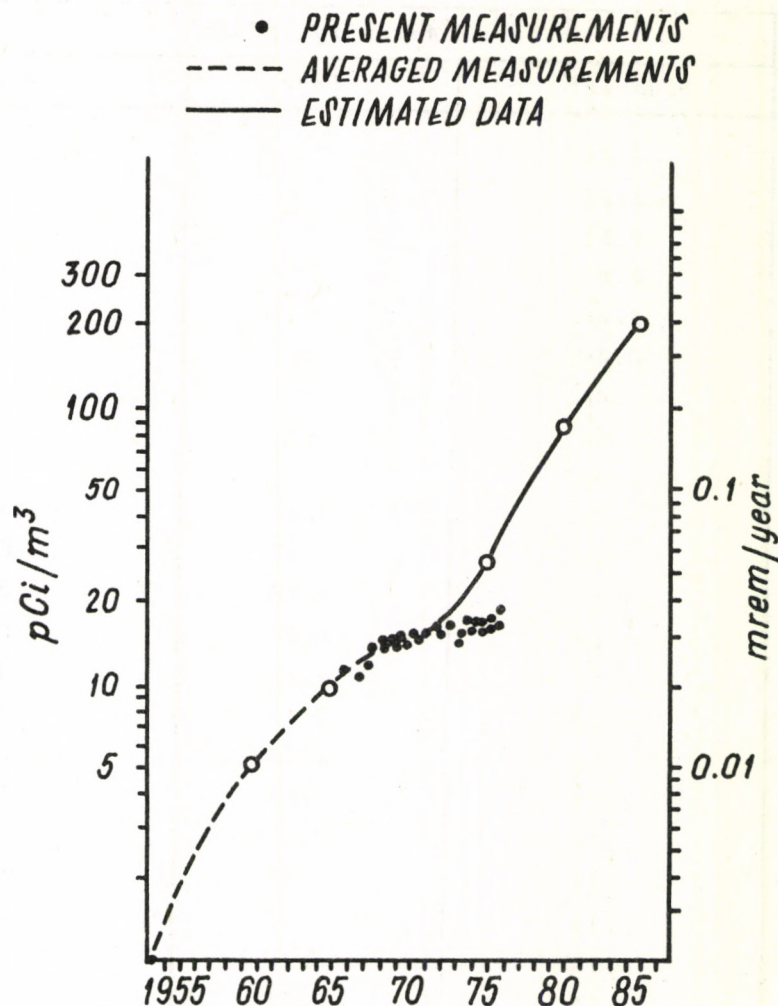
Figure 7

Increase of ^{85}Kr concentration in the atmosphere. Figure demonstrates the data measured in Debrecen between 1966 and 1976 completed with further data published in this field [5]

2. MEASUREMENT OF THE RADIOACTIVE CONTAMINATION OF THE SOIL

Measurement of ^{90}Sr radioactive contamination values of the soil have been carried out in Hungary since 1972. Soil samples taken from 37 sampling sites have been collected once/annum. Sampling sites embrace all over Hungary and demonstrate the main types of its soil /Fig. 8/ [6].
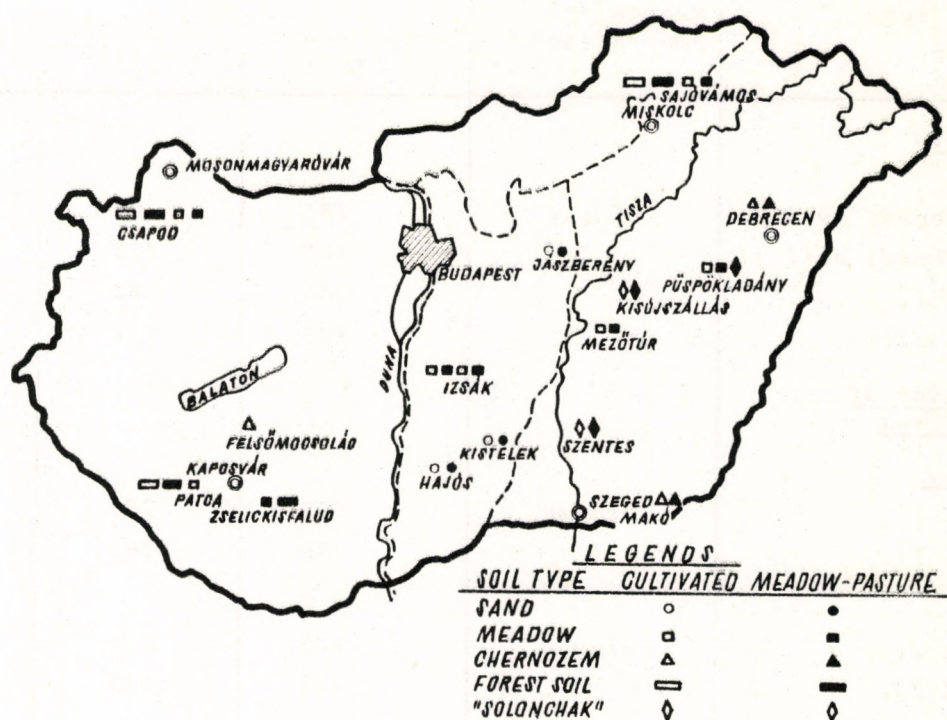
Figure 8

Sampling sites of soils measured from the viewpoint of Sr-90 contamination

In the course of many years measurement, samples taken from the same sampling site originate from both agriculturally cultivated and un-cultivated lands, that is, lands used as a meadow, pasture. Statistical evaluation proved a significantly higher contamination of the meadow, pasture land. From the beginning of the measurement soil sampling sites have been grouped

according to four types of land-units characteristic of Hungary: North Mountain, territory east of the river Tisza, Transdanubia and the Plain between the Danube and Tisza rivers.

Table 6 demonstrates measuring data of the years 1974-1975-1976 according to territorial units and types of soil [6].

Table 6

^{90}Sr *radioactive contamination of soil samples collected in 1974-1975-1976*

Origin and type of the soil	Mechanical composition	^{90}Sr pCi/kg		
		1974	1975	1976
North Mountin				
Sajóvámos				
Chernozem forest soil I.	clayey	382	183	92
Chernozem forest soil II.	"	122	138	74
meadow soil I.	"	165	166	134
meadow soil II.	"	163	137	146
Territory east of the river Tisza				
Püspökladány				
meadow soil I.	clayey	107	188	68
meadow soil II.	"	89	181	76
Mezőtúr				
meadow soil I.	clayey	158	215	93
meadow soil II.	"	208	235	308
Debrecen				
chernozem I.	clayey	104	224	71
Chermozem	sandy	79	272	106
Makó				
meadow chermozem I.	clayey	112	81	10
meadow chermozem II.	sandy clayey	86	70	-
Pankota				
solonchak I.	clayey	105	143	-
solonchak II.	"	159	154	41
Kisujszállás				
meadow solonchak I.	clayey	171	174	59
meadow solonchak II.	"	164	193	63

Origin and type of the soil	Mechanical composition	^{90}Sr pCi/kg		
		1974	1975	1976
Püspökladány				
meadow solonchak II.	clayey	115	200	142
Transdanubia				
Csapod				
black forest soil I.	sandy clayey	123	119	42
black forest soil II.	candy	314	162	140
meadow soil I.	"	145	56	45
meadow soil II.	sandy clayey	346	95	-
Patca				
flooded clayey				
black forest soil I.	clayey	182	138	136
black forest soil II.	"	126	173	67
meadow soil I.	"	136	158	99
Zselickisfalud				
Ramann-black forest soil I.	sandy	192	309	89
medow soil II.	clayey	156	274	202
Felsőmocsoládd				
chermozem I.	"	129	68	89
Plain between the Danube and Tisza rivers				
Izsák				
meadow soil I.	sandy	90	60	67
meadow soil II.	grain sandy	107	80	150
flooded meadow I.	"	106	55	15
flooded meadow II.	"	218	45	62
Hajós				
sandy I.	grain sandy	117	50	37
sandy II.	"	168	34	63
Kistelek				
sandy I.	grain sandy	60	58	64
sandy II.	"	121	119	64
Jászberény				
sandy I.	sandy	137	112	118
sandy II.	"	150	155	149

I. Ploughland

II. Meadow, pasture land

Evaluating the data it can be stated that no outstanding ^{90}Sr contamiation was measured on the territory of Hungary. The maximum value was measured in the territory east of the river Tisza (300 pCi/kg) on a bound meadow soil in 1976.

From the total amount of measuring ^{90}Sr activity exceeded 200 pCi/kg value in case of two samples only. In the majority of cases ^{90}Sr contamination remained below 100 pCi/kg.

The yearly continuous sampling allows to make comparison between the contamination of partical areas and their annual fluctuation. Figure 9 shows the measuring data with respect to the regions between 1972 and 1976, as well as in 5 years' average [6].
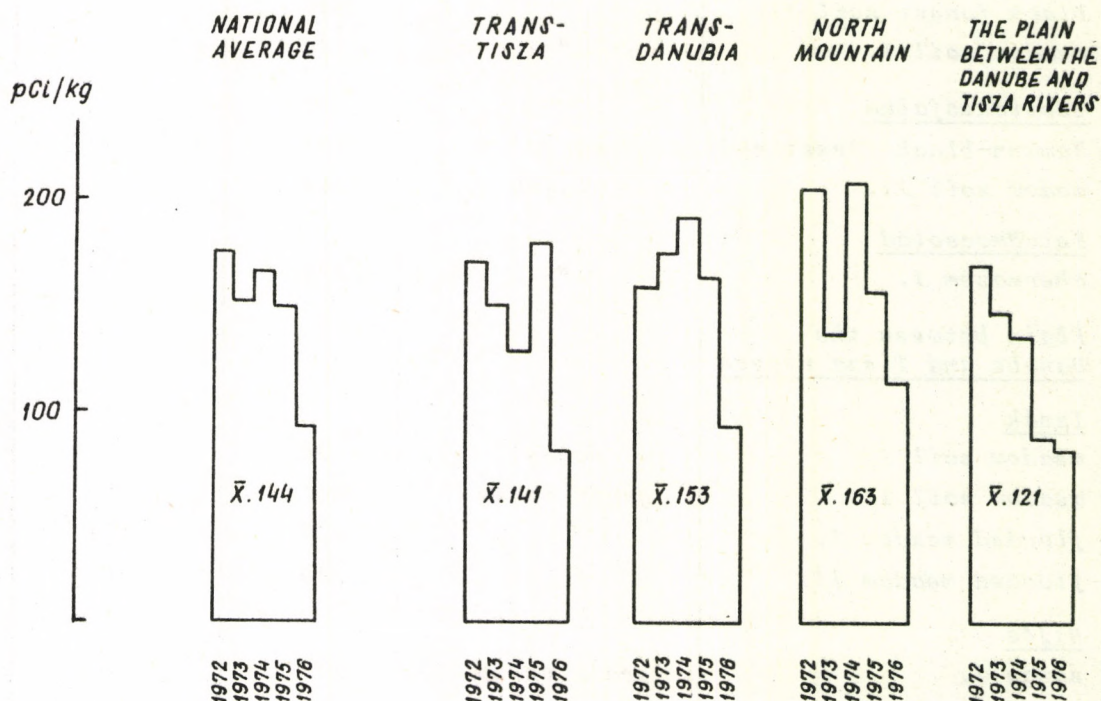
Figure 9

^{90}*Sr contamination of soils measured according to territorial distribution*

According to data, the relative constant values are characteristic of the North Mountain and the territory between the Danube and Tisza rivers. Maximum activities were measured in the territory of the North Mountain. Here, the annual ^{90}Sr contamination was over 100 pCi/kg in general. Glayey soil of heavy mechanical structure is characteristic of this territory. Low contamination values are characteristic of light sandy soils in the plain between the Danube and Tisza rivers. In 1976, an essential change was observed regarding the soil of the territory east of the river Tisza and those of Transdanubia as compared to preliminary years. ^{90}Sr contamination value decreased significantly in both regions. It is especially obvious in Transdanubian territory where, as against to relatively high contamination values observed during the former years, the majority of the measured activity remained below 100 pCi/kg ^{90}Sr, in 1976.

The more extended change of contamination in a territory unit was obserwed in 1975.

Based on statistical evaluations, measurements made on the territory east of the river Tisza showed significant increase while a significant decrease was observed in the territory of the North Mountain and the plain between the Danube and Tisza rivers, in 1975 /Table 7/ [6].

Territory	No. of samples n	^{90}Sr pCi/kg average x	Mean value dispersion S_X	P
Territory east of the river Tisza				
1974	75	127	7.48	P < 0.1%
1975	73	179	7.59	significant
Transdanubia				
1974	55	191	14.7	non-significant
1975	49	161	15.7	
North Mountain				
1974	21	206	19.68	P < 10%
1975	24	156	18.40	significant
Plain between the Danube and Tisza rivers				
1974	54	131	9.98	P < 1%
1975	39	85	11.73	significant

Table 7

Mean values of ^{90}Sr contamination and the significance of their difference measured in 1974-1975

3. MEASUREMENT OF THE RADIOACTIVE POLLUTION OF SURFACE WATERS

The radioactive pollution of the rivers and lakes of Hungary is regularly controlled. The marority of the rivers are running from foreign countries, therefore the sampling sites are mainly at the inflow and exit points, but the large rivers, e.g. the Danube and the Tisza are controlled also at several points with special regard to the drinking water procuring works planted on surface waters.

From another series of measurements the contamination of some of the larger rivers of Hungary and the Lake Balaton is presented between 1965 and 1976 in Table 8 [7].

Year	Danube	Rába	Dráva	Tisza	Sajó	Balaton
1965	13	36	16	14	40	11
1966	9	29	14	15	34	9
1967	9	30	13	16	50	9
1968	3	44	13	14	40	7
1969	4	37	11	14	13	6
1970	9	23	15	15	23	15
1971	8	29	17	5	18	16
1972	8	29	20	11	14	10
1973	10	25	18	8	14	10
1974	9	34	27	7	7	12
1975	10	22	21	13	6	14
1976	10	22	5	15	10	11

Table 8

*The average gross beta-activity of surface waters in Hungary
in pCi/l [7]*

The activity of ^3H, ^{90}Sr, ^{137}Cs in the Danube at Budapest in 1976 is presented in Table 9 [7]

The gross beta-activity of the Danube at Budapest and at the exit from the country and its seasonal distribution in 1976 is demonstrated in Table 10 [7].

Month		I.	II.	III.	IV.	V.	VI.	VII.	VIII.	IX.	X.	XI.	XII.
H-3	Min.	116	109	108	110	107	97	116	116	109	104	103	97
TU	Max.	138	130	143	142	136	107	145	132	128	128	124	119
	Average	123	120	123	124	124	102	129	125	117	115	112	106
Cs-137	Min.	1.6	1.6	1.3	0.9	0.9	2.3	0.2	0.3	0.2	1.6	0.3	0.7
pCi/l	Max.	2.3	2.9	2.6	1.8	2.4	2.4	2.8	2.0	2.9	2.4	3.0	3.3
	Average	2.0	2.4	2.0	1.3	1.9	2.3	1.8	1.2	1.6	1.9	1.5	2.0
Sr-90	Min.	1.2	0.9	1.2	0.0	1.2	0.0	0.3	0.8	1.2	0.5	0.1	0.4
pCi/l	Max.	1.5	2.5	2.9	3.5	2.4	1.9	0.7	3.2	2.9	2.8	2.0	4.3
	Average	1.3	1.7	2.5	1.4	1.7	0.8	0.5	1.6	2.0	1.3	1.2	2.1

Table 9

^3H, ^{137}Cs and ^{90}Sr activity of the Danube at Budapest 1976 [7]

Month		I.	II.	III.	IV.	V.	VI.	VII.	VIII.	IX.	X.	XI.	XII.
	Min.	5	8	6	9	7	5	5	2	3	3	4	3
At Budapest	Max.	10	12	20	15	25	16	20	6	7	7	11	11
	Average	8	8	11	12	12	10	9	4	5	5	6	7
At the	Min.	2	1	1	3	2	2	3	2	3	3	5	3
exit point	Max.	11	13	16	18	9	12	10	6	7	7	11	11
	Average	5	5	7	7	4	6	5	4	5	5	6	7

Table 10

The gross beta-activity of the Danube in 1976 [pCi/l] [7]

4. MEASUREMENT OF RADIOACTIVE CONTAMINATION OF MATERIALS OF VEGETAL AND ANIMAL ORIGIN AND FOODSTUFFS

The determination of the activity of samples includes besides the continuous measurements of the radioactive contamination level and the analysis of the trends of the changes the study of the uptake and the way of incorporation of the radioactive elements.

The choice of the sampling sites was performed on the base of the territorial properties /see Fig. 10 and 11/[8]. Their equal distribution makes possible to get adequate information from the whole country.

Figure 10

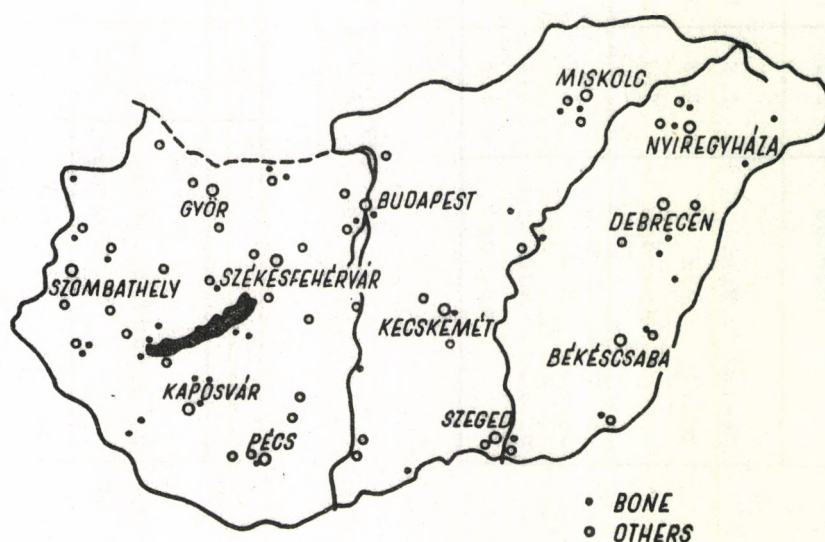*Sampling sites for vegetables and milk in Hungary [8]*

Figure 11

Sampling sites for bone and others in Hungary [8]

Supplementing the data it has to be emphasized that in addition to the evaluation of the contamination of the individual samples, the statistical evaluation of the interaction of the sampling sites and times was also carried out.

Measurement of Vegetables

Among the products of vegetal origin as representative samples, spinach, lettuce and oxalis /comon sorrel/ were studied. These vegetables represent some indicators among the regularly comsumed group of products and they supply reliable information concerning the quantity of material taken up either from the fall-out or from the soil /with special regard to metal ions/.

The data obtained from systematic investigations were evaluated from three main sampling regions: I. Transdanubia; II. the plain between the Danube and Tisza rivers; III. Trans-Tisza region.

Annual average data-values concerning spinach, lettuce and oxalis are shown in Tables 11-13 [8].

Time of sampling		Area of sampling		
		I. Transdanubia	II. Plain Region between Danube and Tisza	III. Trans-Tisza region
1959		7.1	3.1	5.2
1960,	spring	1.4	2.7	1.6
	autumn	0.9	4.6	1.1
1961,	spring	0.6	0.9	0.4
	autumn	1.0	1.0	0.5
1962,	spring	26.2	10.8	12.6
	autumn	6.1	8.8	9.6
1963,	spring	35.8	102.9	34.6
	autumn	13.3	19.8	12.7
1964,	spring	10.5	7.3	4.3
	autumn	2.0	2.2	2.5
1965,	spring	4.0	4.0	4.2
	autumn	1.9	1.4	2.4
1966,	spring	1.5	1.7	1.2
	autumn	2.4	0.0	1.3
1967,	spring	2.1	2.0	1.8
	autumn	2.5	5.4	1.4
1968		3.0	4.1	2.3
1969		3.0	4.1	2.4
1970		3.8	3.7	4.0
1971		3.5	4.1	3.0
1972		4.5	5.1	2.9
1973		2.2	3.3	1.2
1974		4.0	2.9	3.4
1975		2.6	2.3	2.3
1976		13.2	8.2	5.9

Table 11

Activity of metal ion fraction indicating the radioactive contamination of spinach, pCi/g dry substance [8]

Time of sampling		Area of sampling		
		I. Transdanubia	II. Plain Region between Danube and Tissa	III. Trans-Tissa region
1959		2.9	1.8	3.3
1960,	spring	2.3	1.5	1.6
	autumn	3.3	2.0	3.1
1961,	spring	1.2	1.1	1.4
	autumn	1.4	1.9	2.0
1962,	spring	19.8	5.7	9.5
	autumn	7.0	13.3	6.7
1963,	spring	19.5	22.4	19.6
	autumn	10.4	23.3	10.9
1964,	spring	3.5	6.4	7.4
	autumn	1.7	3.9	2.4
1965,	spring	3.2	5.3	3.6
	autumn	1.7	2.8	1.9
1966,	spring	4.6	5.2	3.1
	autumn	2.9	3.1	1.5
1967,	spring	4.6	2.5	3.1
	autumn	2.6	6.0	3.7
1968		4.7	2.5	2.5
1969		3.7	4.1	2.4
1970		3.5	3.5	3.7
1971		4.0	2.2	3.3
1972		4.7	3.5	2.7
1973		3.2	3.3	1.5
1974		3.7	2.7	2.6
1975		2.2	2.2	2.3
1976		12.4	6.1	5.1

Table 12

Activity of metal ion fraction indicating the radioactive contamination of lettuce pCi/g dry substance /8/

Time of sampling		Area of sampling		
		I. Transdanubia	II. Plain Region between Danube and Tissa	III. Trans-Tissa region
1959		3.6	4.1	1.6
1960,	spring	2.0	1.8	1.9
	autumn	1.1	1.8	0.9
1961,	spring	1.0	1.3	0.9
	autumn	1.8	2.5	1.2
1962,	spring	22.1	12.0	10.2
	autumn	6.3	21.1	9.6
1963,	spring	21.7	21.5	15.3
	autumn	17.4	19.8	17.0
1964,	spring	9.0	8.5	7.1
	autumn	2.1	4.6	3.0
1965,	spring	4.2	4.2	3.7
	autumn	1.6	1.5	2.4
1966,	spring	1.1	2.6	1.6
	autumn	1.6	1.3	2.2
1967,	spring	2.2	2.0	1.2
	autumn	2.8	1.7	2.8
1968		3.0	3.9	2.8
1969		3.5	3.7	2.4
1970		4.8	3.1	5.2
1971		3.3	4.2	3.4
1972		3.8	3.2	2.8
1973		2.3	2.9	1.1
1974		3.5	2.2	2.5
1975		2.3	2.2	1.7
1976		12.3	6.2	4.6

Table 13

Activity of metal ion fraction indicating the radioactive contamination of oxalis, pCi/g dry substance /8/

The activity of the metal ion fraction is the result of the activity of radioactive elements coprecipitated with ^{90}Sr.

Results of variance analysis may be summed up as follow:

/1/ From spring, 1962 to the end of 1964 the contamination of each sample was significantly higher than in the previous and during the subsequent years while a significantly higher value was measured again in 1976;

/2/ Based on the analysis according to the regional distribution, no significant differences between the various sampling sites were observed;

/3/ Neither specific control examinations nor results of regular measurements evidenced higher contamination to be qualified as particularly contaminated or to be submitted for more careful control from the viewpoint of the fall-out or the production.

Measurement of Fodder and Crops

Results obtained with fodder examinations made between 1965 and 1976 are demonstrated in Table 14 /8/.

Year	Hay			Silage		
	Average	Maximum	Minimum	Average	Maximum	Minimum
1965	5.6	8.3	3.9	5.6	13.8	0.5
1966	3.4	5.5	1.8	3.3	4.2	2.5
1967	2.6	4.9	1.4	2.7	5.4	0.8
1968	2.4	5.1	0.6	2.3	6.0	0.3
1969	3.3	3.8	0.4	3.2	8.7	0.4
1970	3.7	7.5	1.4	3.3	-	-
1971	5.7	8.2	0.7	5.5	15.4	2.4
1972	3.4	-	-	3.4	-	-
1973	2.3	-	-	3.1	-	-
1974	3.0	-	-	3.0	-	-
1975	2.6	-	-	-	-	-
1976	1.8	-	-	-	-	-

Table 14

Radioactive contamination of fodder in the years 1965-1976. Activity of metal ion fraction in pCi/g dry substance /8/

Annual averages of both hay and silage are rather uniform and their activity is ranging within the same order of magnitude as that of the indicator vegetables, i.e. spinach, lettuce, oxalis /see Tables 11-13/.

The activity of the metal ion fraction in wheat between 1971 and 1976 is summarized in Table 15 /8/.

Sample	Total activity	Activity of the metal ion fraction	^{40}K activity
Wheat			
1971	3.9	0.2	3.3
1972	3.6	0.1	3.1
1974	3.8	0.3	3.5
1975	4.1	0.3	3.6
1976	4.3	0.2	3.6

Table 15

Radioactive contamination of crops in the years 1971-1976 in pCi/g dry substance /8/

Measurement of Tobacco

Year	Total activity	Activity of the metal ion fraction	^{40}K activity
1971	42.52	9.94	24.45
1972	38.00	5.90	26.30
1973	31.20	4.20	24.60
1974	38.60	5.50	30.30
1975	30.90	4.80	23.80
1976	35.30	4.70	29.60

Table 16

Radioactive contamination of tobacco, in the years 1971-1976 in pCi/g dry substance /8/

Measurement of Milk

Table 17 /17,18/ demonstrates the results obtained with the representative measurements of milk samples /8/.

The radioactive contamination is constant enough and the relatively slight differences may be deducted from the radioactivity of the consumed fodder and forages of various composition.

Jear	Activity of metal ion fraction
1960	4.9
1961	2.4
1962	2.8
1963	6.9
1964	2.2
1965	3.2
1966	2.1
1967	2.3
1968	1.9
1969	2.0
1970	2.2
1971	2.2
1972	2.4
1973	2.4
1974	2.6
1975	2.0
1976	1.5

Table 17

Activity of the metal ion fraction of milk in the years
1960-1976 in pCi/100 g milk [8]

Measurement of Fish

The measurement of fish samples of various origin supplies information concerning the degree of pollution occuring under river- and lake-keeping circumstances; see Table 18 [8]. It is obvious that the activity of the metal ion fraction characteristic of the artificial radioactivity is higher in fish bones than in muscles.

Sample /year/	Bone			Muscles		
	Total activity	Activity of metal ion fraction	^{40}K act.	Total activity	Activity of metal ion fract.	^{40}K act.
Various fish samples						
1973	9.1	5.4		10.6	0.3	8.8
1974	8.3	4.0		33.3	1.0	31.3
1975	7.3	4.6		11.3	0.2	9.8
1976	6.3	2.4	3.7	11.1	0.2	9.5

Table 18

Radioactive contamination of fish in the years 1973-1976
in pCi/g dry substance [8]

Radioactive Contamination of Animal Bones

Animal bones were studied for the determination of the incorporation of various radioactive elements.

Contamination data of bone samples of various origin are presented in Table 19 [8].

Sample	Year	Total activity, pCi/g bone	Metal ion fraction, pCi/g bone	^{90}Sr activity pCi/g Ca
Calf	1971	3.3	2.8	–
	1972	3.1	2.5	23.2
	1973	2.6	1.6	–
	1974	2,8	2.2	–
Young cattle metacarpus	1971	3.2	6.2	
	1972	4.8	3.6	
	1973	4.7	3.5	
	1974	3.1	2.2	
	1975	4.9	2.5	
	1976	1.8	1.7	
Sheep bone	1972	6.0	4.5	
	1973	4.8	3.4	
	1974	7.4	6.5	
	1975	4.8	2.5	
	1976	1.7	1.7	
Game bone	1972	8.3	7.2	
	1973	10.3	7.8	
	1974	7.8	5.4	
	1975	–	8.8	

Table 19

Activity of bone samples of various origin in the years 1971-1976 [8]

It can be stated that the artificial radioactive contamination may be connected with the age, species as well as the different circumstances of keeping, feeding and grazing of the animals.

5. INCORPORATION STUDIES

^{137}Cs incorporation can be detected in the human body as a result of air pollution due to nuclear explosions. Yearly doses based upon whole-body counting measurements between 1964 and 1976 are demonstrated in <u>Fig. 12</u>. According to the calculations 86.1 pCi ^{137}Cs/kg body weight would result 1 mrem/year dose burden [9].
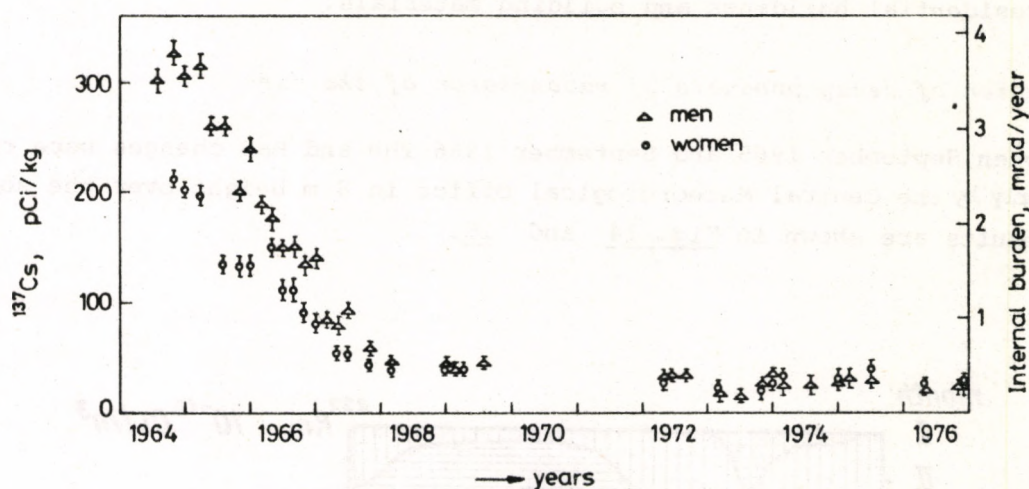

<u>Figure 12</u>

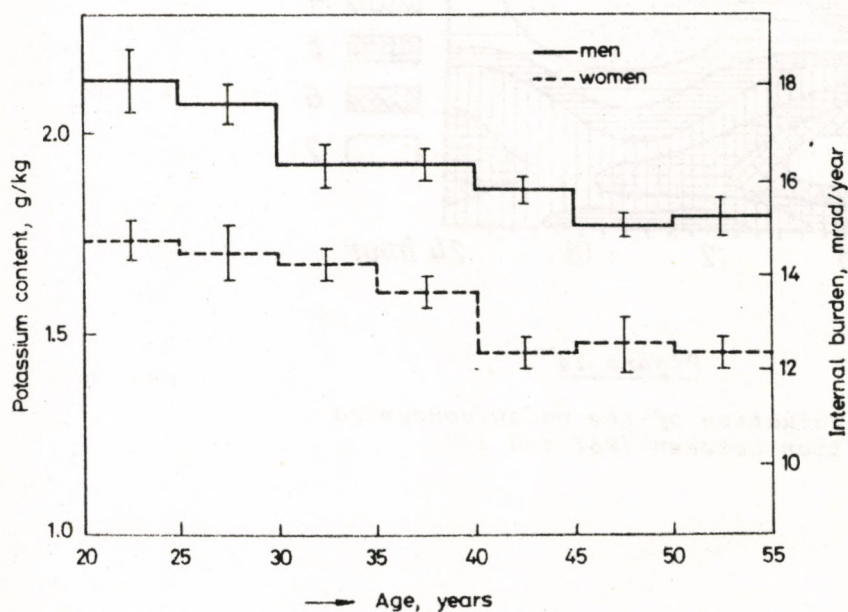*Whole-body counting measurements between 1964 and 1976*

<u>Figure 13</u> shows the potassium content in male and female bodies as well as the alteration of radiation burden in the function of the age [9].

<u>Figure 13</u>

Potassium content in male and female bodies

6. NATURAL RADIOACTIVITY

The investigation of artificial radioactive contamination getting to the environment turned the attention on an ever increasing manner to natural radionuclides to be observed in the environment as well. Its measuring was necessitated partly by the investigation of the radioactive aerosol content of the air, and partly by getting known of the natural radiation burden of the population of such an origin. Accordingly, investigations related to several parts, such as the measurement of radon and toron decay products, natural radioactive material content of the soil, polonium-210 content of tobacco and the investigation of residential buildings and building materials.

Investigation of decay products of radon-toron of the air

Between September 1965 and September 1966 ThB and RaB changes were registered hourly by the Central Meteorological Office in 8 m height over the soil level, results are shown in Fig. 14 and 15.
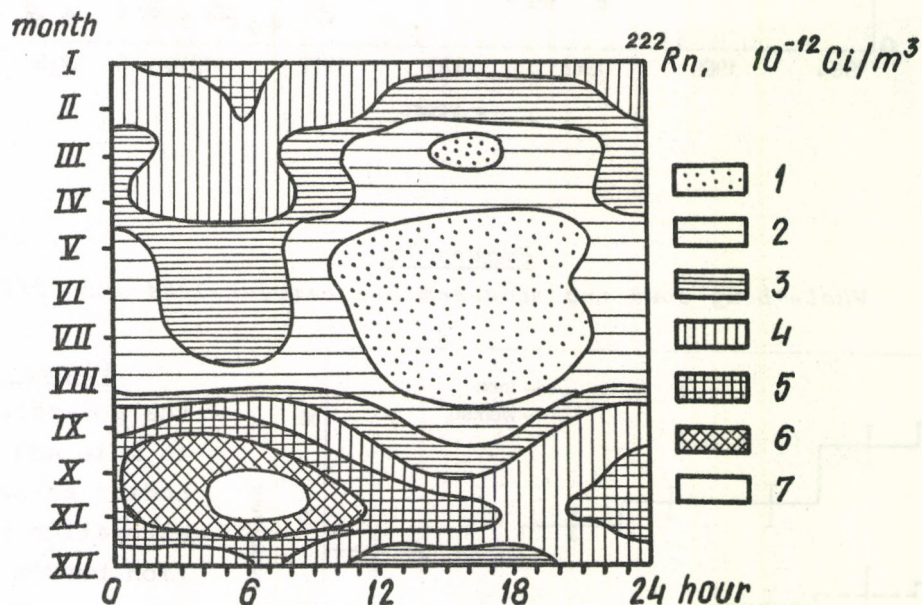

Figure 14

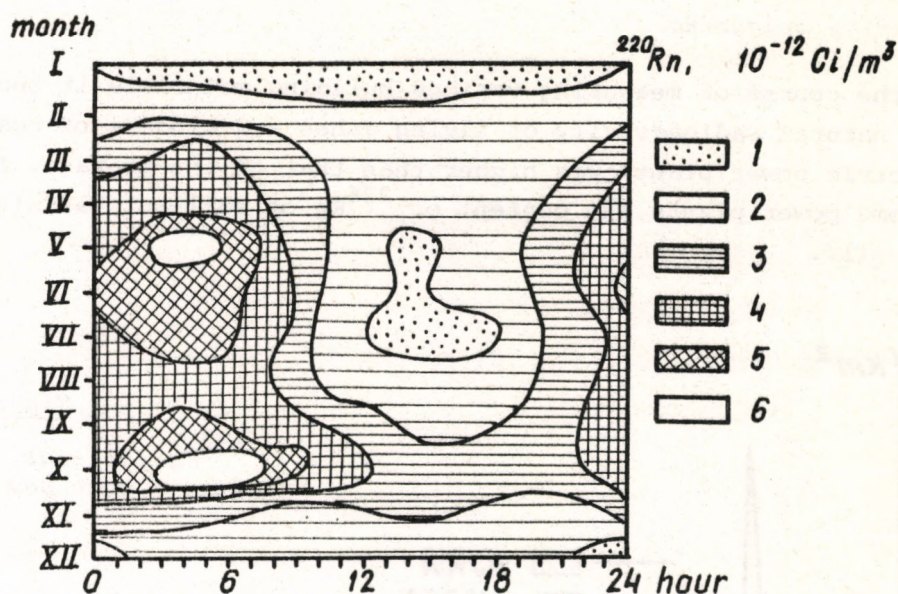*Fluctuation of the radon concentration between 1965 and 1966*

Figure 15

*Fluctuation of the toron concentration
between 1965 and 1966*

As for ThB the values per hour ranged between 50-150% as compared to the mean value and 67-144% with respect to RaB. Monthly mean values of ThB/RaB rate are demonstrated in Table 20 [10].

Months		Months	
January	0.0066	July	0.0741
February	0.0175	August	0.0760
March	0.0401	September	0.0396
April	0.0403	October	0.0336
May	0.0890	November	0.0150
June	0.0882	December	0.0134
		Annual mean	0.0444

Table 20

ThB/RaB rate monthly mean values

It can be stated that there is a significant difference to be observed regarding both ThB and RaB absolute values and their ratio to each other depending on the season and hour of the day.

34

Soil activity measuring

In the course of measuring various building materials it became obvious that the natural radioactivity of flying ashes and cinders of coal used in some electric power plants was higher than that of the average. In the vicinity of some power plants the content of ^{226}Ra of fall-out was significant /Fig. 16/ [11].
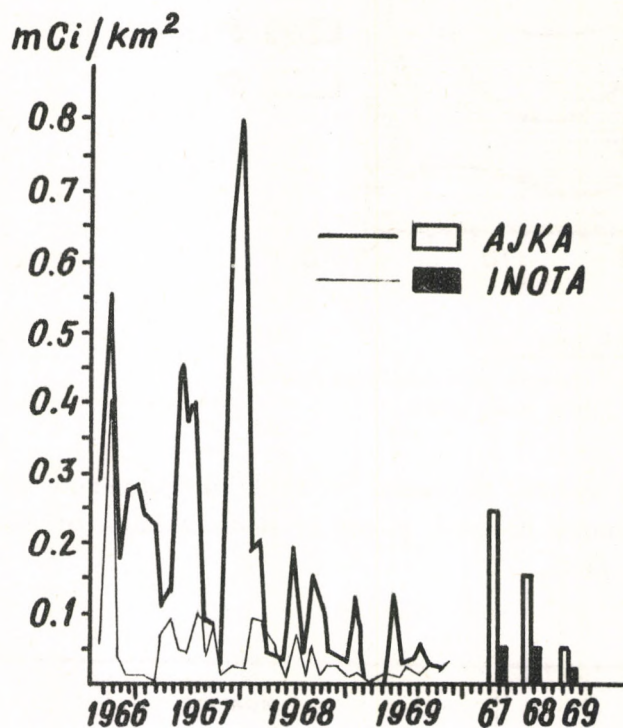

Figure 16

^{226}*Ra fall-out in the vicinity of some power plants*

Measurements were made in the vicinity of power plants, in some other places taken as a control and in Budapest where cinders were used of such an exposed origin. Results are shown in Table 21 and 22 [12].

Sampling site	Character number of samples		pCi/g
Ajka	flying ashes	3	229.4-252.4
Tatabánya	flying ashes	5	144.3-209.6
Inota	flying ashes	3	58.3- 81.1
Inota	cinders	3	53.3- 67.2
Tatabánya	mixed, slate, waste tip	2	23.0- 26.8
Tatabánya	gravel substitutive	2	75.7- 86.0
Mátra-region /Mátra-vidék/	gravel substitutive	1	33.1

Table 21

α-activity of flying ashes, cinders and gravel substitutive samples pCi/g

Measurement of polonium-210 content of tobacco

For the determination of radioactive material content of cigarette-smoke getting into the organism and attacking the bronchi first of all, the polonium-210 content of tobacco was measured. Measurements were made with tobacco purchased between November 1969 and December 1970, thus cigarettes have been prepared from tobaccoleaves grown one or two years earlier, very probable /Table 22/ [13].

	pCi/g
Filtered	0.58
Unfiltered	0.55

Table 22
^{210}Po content of cigarette tobacco [13]

Radioactive measurement of building materials

For the evaluation of the natural radiation burden of the population, it is necessary to know the radioactive nuclide content of the building material of res dential buildings. Measurements have been carried out for many years. Results are summed up in Tables 23 and 24. It is evident that with the exception of some flying ash and cinder samples the natural activity of the usual building materials is of the same or similar order of magnitude [14].

Building material and properties	Concentration /pCi/g/			Emanation factor for radon /%/ released Rn/total Rn
	^{228}Th	^{226}Ra	^{40}K	
Concrete, precast slabs fabricated in Budapest plant /radium-free Danube ballast very porous/	0.3	0.2	7.0	47
Red bric /loose, with fissures, humid fabricated in Pécs/	1.7	1.5	16.0	12
Adobe brick a./ from demolition in Pécs	1.6	1.8	23.0	35
b./ new, from Tápió-györgye	1.7	1.2	16.0	37
c./ old, very compact from Lágymányos	1.1	1.5	12.0	16

Table 23
^{226}Ra, ^{228}Th and ^{40}K activity concentrations in different building materials

| No. | Denomination of the sample | Weight of the sample, g | Density, g/cm^3 | Fissures | Number of | | Effective ^{226}Ra content fCi/g | | |
					samp-les	Rn-meas-urements	min.	max.	average
1	Concrete trial blocks	19 000	2.4	no	28	34	80	110	100
1/a	Danubian ballast a	14 200	1.6	no	3	2	−	−	9
1/b	"Keramzit" balls b	3000	1.1	yes	1	1	−	−	50
2	Blast furnace slag concrete	13 000	2.0	some	2	2	−	−	295
3	Trial blocks of concrete made with fly-ash c	7300-15400	1.2	no	11	32	280	3290	750
4	Red bricks /Pécs/	10 700	1.6	yes	4	5	185	191	190
5	Reddish-yellow bricks	9000	1.6	yes	4	8	130	160	140
6	Bricks for curtain-walls	2800-12 000	1.5	no	14	14	40	140	80
7	Adobe bricks	4800-10 330	$1.2; 1.7^d$	yes	4	5	250^d	670	490
8	Interior paving-blocks and floor-tiles	6400-12 000	2.3	no	6	8	40	150	80
9	Exterior paving blocks	650-6400	1.9	no	3	4	−	−	50

Remarks to Table 2: a/ as aggregate for concrete in 7 cases
b/ as additive agent for thermal insulation in concrete
c/ used for backfilling purposes only /800 kg fly-ash, and 160 kg cement, per m^3 concrete/
d/ 250 fCi/g at 1.7 g/cm^3 density

Table 24
The effective ^{226}Ra content of the building /backfilling/ materials

Natural radioactivity of the air of residential building indoors

The measurement of building materials of various origin arose the probability that even in Hungary the natural radioactivity of indoor air or resedential buildings created according to various methods of building and of various materials would be different. This presupposition was confirmed by informative initial measurements /Table 26/ [15].

Premises	*Ci/1*
Buildings of fly ash and of slag	$1,4 \cdot 10^{-10}$
Closed brick buildings	$8,9 \cdot 10^{-13}$
Inhabited brick buildings	$3,8 \cdot 10^{-13}$
Outdoors	$8,9 \cdot 10^{-14}$

Table 25

Radon concentration in the air buildings [15]

Thereafter, the full measurement took place in different regions of the country in houses made of the most commonly used building materials at various floor levels above the soil level for 1970. Results are demonstrated in Figs. 17 and 18 [16].
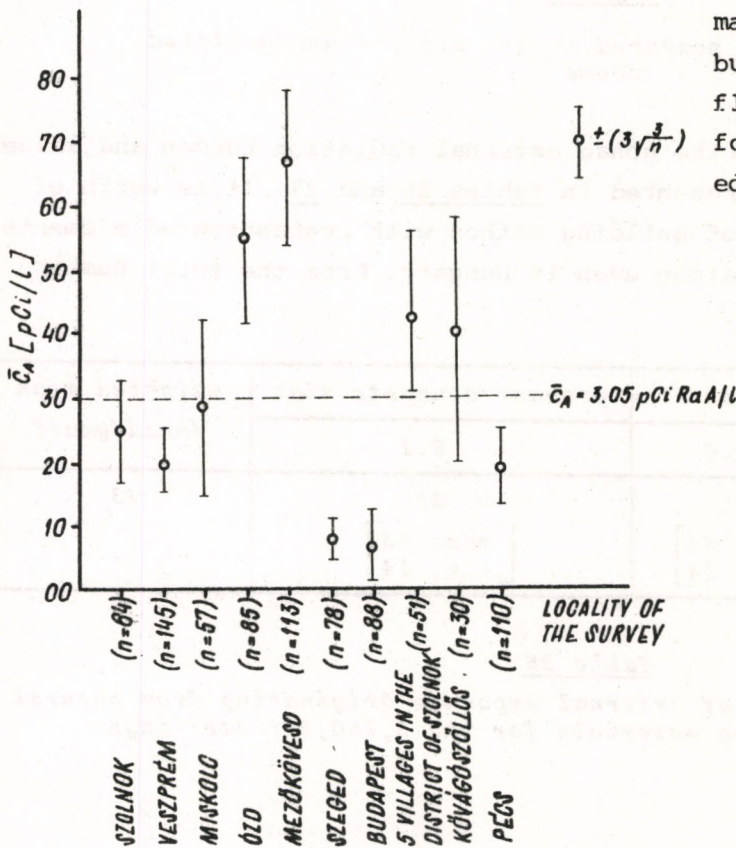

Fig. 17

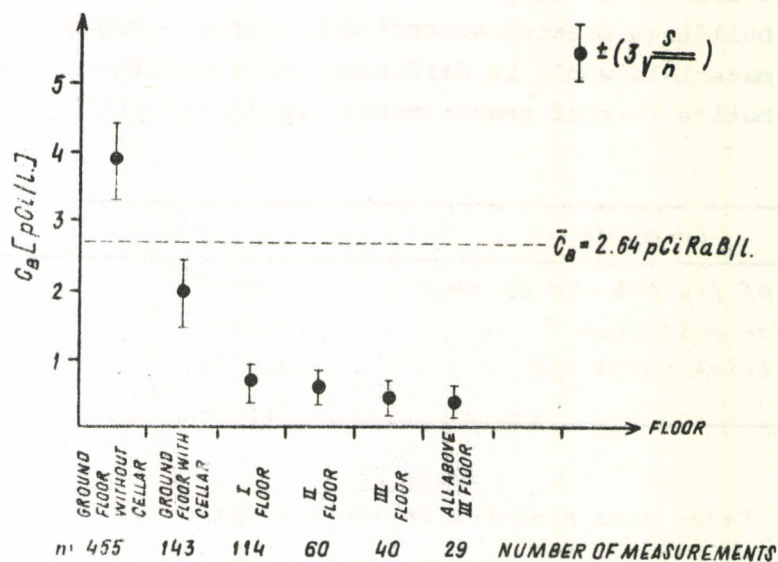*RaA concentrations measured in the air of unventilated rooms*

Fig. 18

RaB concentrations measured in the air for unventilated rooms

Based on measuring results the gonad external radiation burden and values of bronchial dose burden are presented in Tables 26 and 27. It is worth of mentioning that the spreading of building method with prefabricated elements would bring essential changes along even in Hungary. From the total number

walls	adobe	brick	precast concrete slab	weighted mean /mrad/year/
%	57.5	34.4	8.1	
gonads /mrem/year	33 $\begin{bmatrix} max. & 60 \\ min. & 10 \end{bmatrix}$	32 $\begin{bmatrix} max. & 63 \\ min. & 14 \end{bmatrix}$	38 $\begin{bmatrix} max. & 63 \\ min. & 14 \end{bmatrix}$	33

Table 26

The dose equivalent of external exposure originating from natural sources of building materials for the 3,150,267 dwellings

walls	adobe	brick	precast concrete slab	weighted mean /mrad/year/
%	57.5	34.4	8.1	
bronchial /mrad/year/	1058	318	150	730

Table 27

The bronchial dose rate from radon-daughters in 540 dwellings of Hungary

of residential buildings of 245,772 50% has been made of prefabricated elements, 40% of brick and 10% of adobe, build between 1970 and 1972. This fact modifies the external dose burden to 34 mrem/year. However, the bronchial dose lowers to 308 mrad/year taking less than the half of the former value [17].

7. SUMMARY

The detailed study of the data shows that the amount of the radioactive pollution /contamination/ is comparable with that of those regions of similar geographical and meteorological conditions as Hungary and are situated between 46 and 49 degress of latitude with 500-600 mm annual amount of precipitation.

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