Environmental Radioactivity in Hungary

Bulletin No. 1

EDITED BY
J. KOVÁCS AND
T. PREDMERSZKY

BUDAPEST 1975

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C O N T E N T S

PREFACE

During the ninteen-fifties a high number of atmospheric and underwater atomic bomb explosions were performed, inducing the ever increasing radioactive pollution of the atmosphere. This indicated, in the first place, the beginning of systematic examinations of the atmospheric radioactive contamination in several countries, so in Hungary too.

Later on the measurements were extended also to the water surfaces, the soil, the foodstuffs, e.g. the vegetal and animal substances forming the most important elements of food and even to man concerning their radioactivity. The increasing number of large nuclear establishments rendered this demand even more important.

Since the spreading of the contamination in the atmosphere or in running waters cannot be restricted by frontiers, it seemed advisable to exchange mutual information concerning the values of radioactive contamination in the individual countries. Thus, already during the ninteen-fifties the first periodical national publications containing pertinent information came into existence.

In Hungary, although some of the authors published from time to time the results of their measurements in special journals and at conferences, no summarising documentation appeared. Our present publication the "Environmental Radioactivity in Hungary Bulletin" was prepared with the intention to meet this need. According to our plans, it will be followed by a series of new publications in the future.

Prof. Dr. L. Bozóky

Investigation of Environmental Radioactive Contamination in Hungary

The plan of the investigation of environmental radioactive contamination in Hungary was prepared in the early fifties and included all those sampling sites, which are suited within various climatic zones and cultural regions for the determination of the degree of radioactive contamination.

The study of the radioactive contamination of the predictable soil surface accumulation and of the products of vegetal and animal origin coming from various regions of the country as well as the analysis of their differences were suited for the determination of the effect of the factors influencing the contamination.

Among the radioactive contamination studies first the sampling and measurements for assessing atmospheric contamination was started at the beginning of the fifties. Systematic determination of radioactive contamination appearing in vegetal and animal samples has been performed only from the year 1959.

Parallel to the assignement of national sampling and control sites, laboratory methodological studies were performed in order to elaborate the methods adequately exact and suited also for routine investigations /and correspond essentially to the methods described in the literature/.

The following chapters of this paper contain the considerable part of available informations about Hungarian radioactive contamination studies. These may be considered for the summarising evaluation of the systematic monitoring, carried out since about 15-20 years.

1. STUDIES ON ATMOSPHERIC RADIOACTIVE CONTAMINATION

The first systematic atmospheric pollution studies were carried out in 1952. During the early period - in Hungary as well as all over the world - the degree, changes and spreading of radioactive contamination of artificial origin was examined.

The atmospheric radioactive contamination is present partly in gaseous form, partly in aerosols. From the aspect of its composition the atmospheric radioactivity can be divided into two large groups. Accordingly the individual fractions were examined:

/1/ Radioactive substances of natural origin:

/a/ gaseous products of natural radioactive substances, e.g. Rn, Tn as well as their decay products;

/b/ the cosmogenic radioactive substances, originating from the interaction of cosmic radiation with the gases of the atmosphere of the earth, e.g. ^{14}C, ^{3}H, ^{7}Be.

/2/ Atmospheric contamination produced by human activities:

/a/ fission and activation products produced by nuclear explosion tests;

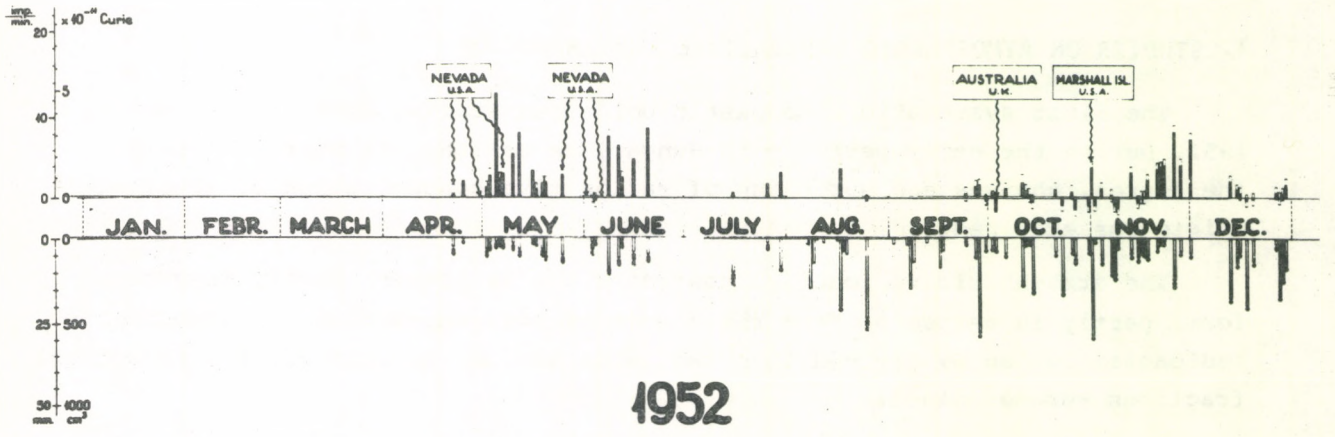
/b/ various fission products released in course of reactor operation and elaboration of nuclear fuel elements.

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 1957 as well as 1952 - 1969 are shown in Fig. 1 [1] (based on the results of the investigations there was a possibility for the approximate determination of the time of the atomic bomb explosion tests in the beginning) and Fig. 2 [2], respectively.

Figure 1

Beta activity of fission products in the atmospheric precipitation in Debrecen, Hungary, during 1952 and 1973. Ordinate upwards, right: activity in 10^{-11} Curie units, corrected for the geometry of the counting equipment. Ordinate upwards, left: activity observed in cpm, both reduced to 1/50 m^2 ombrometer surface. Ordinate downwards, right: one day's rainfall in ml water volume collected by an exposed area of 1/50 m^2. Ordinate downwards, left: one day's rainfall in mm [1]



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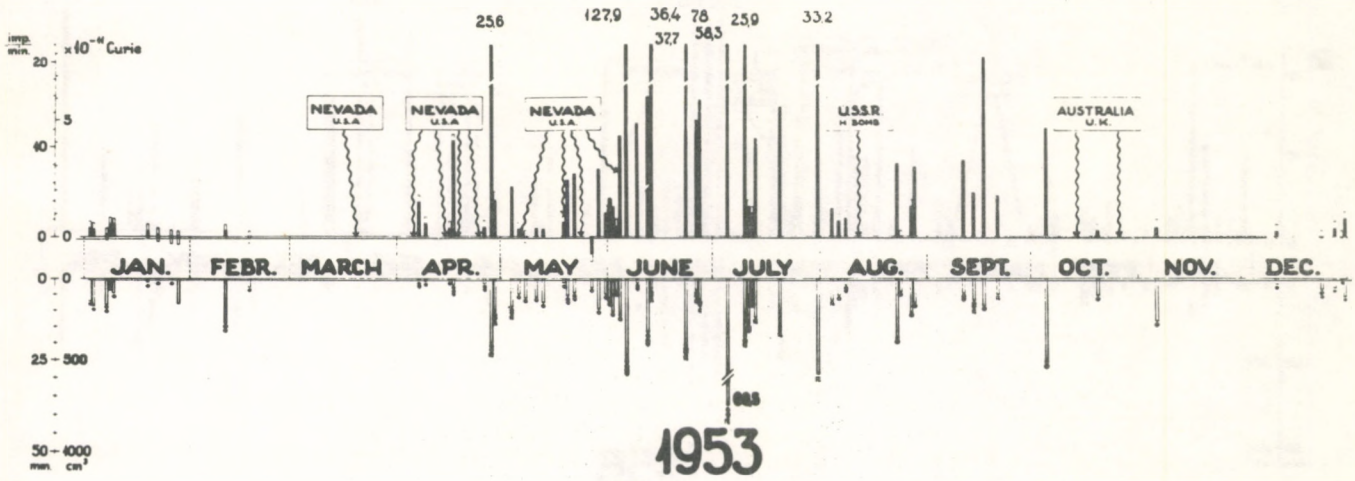
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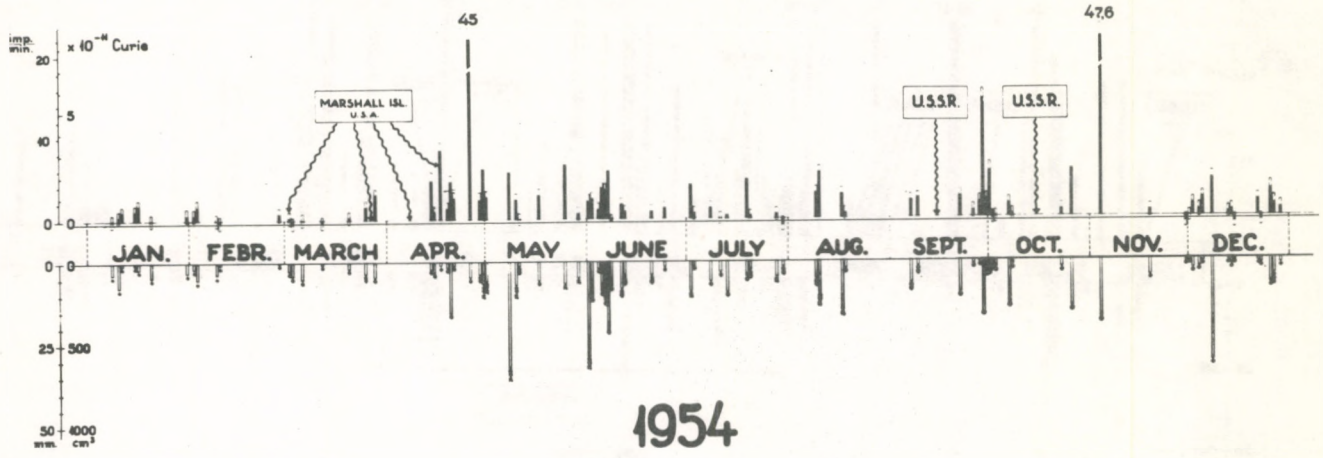
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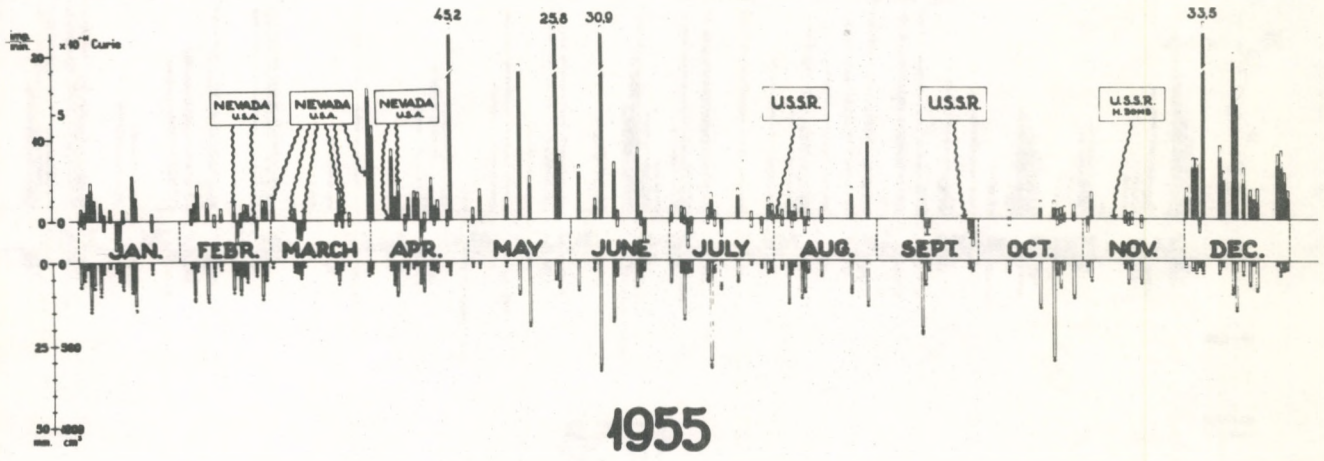
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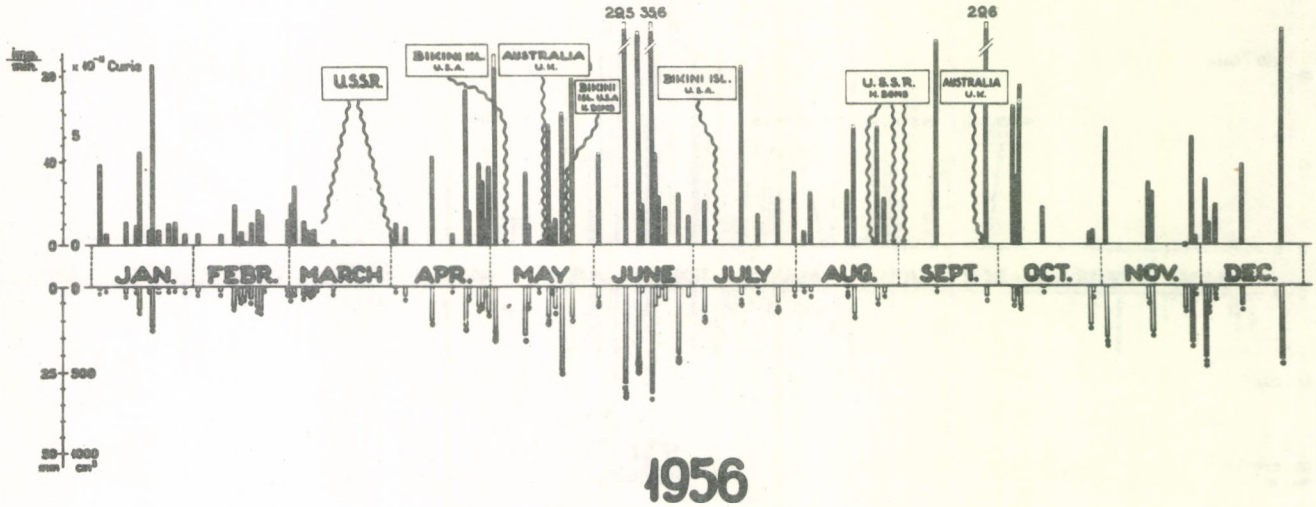

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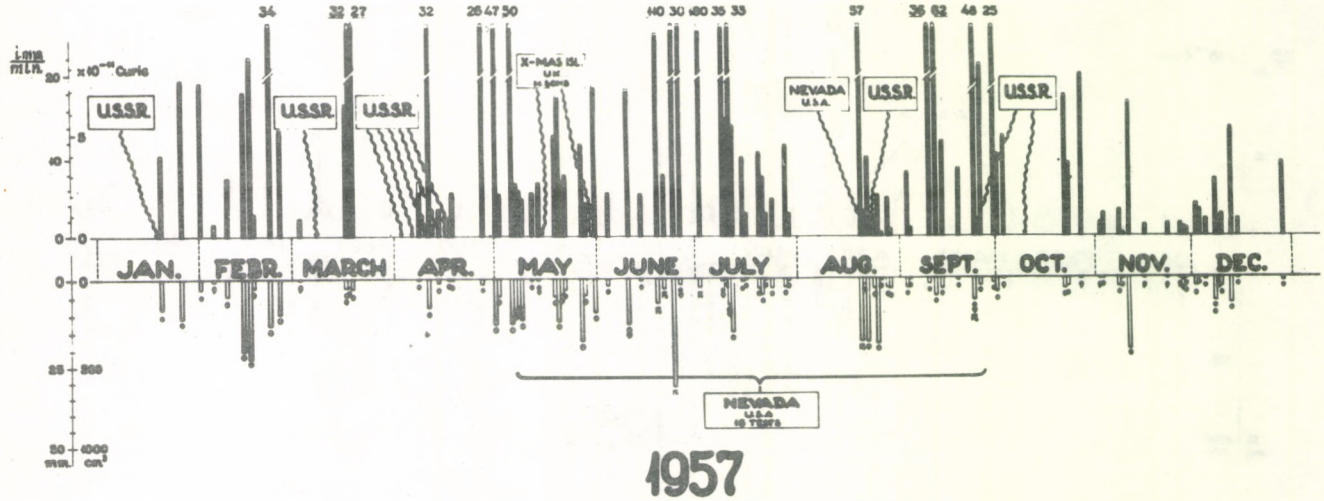

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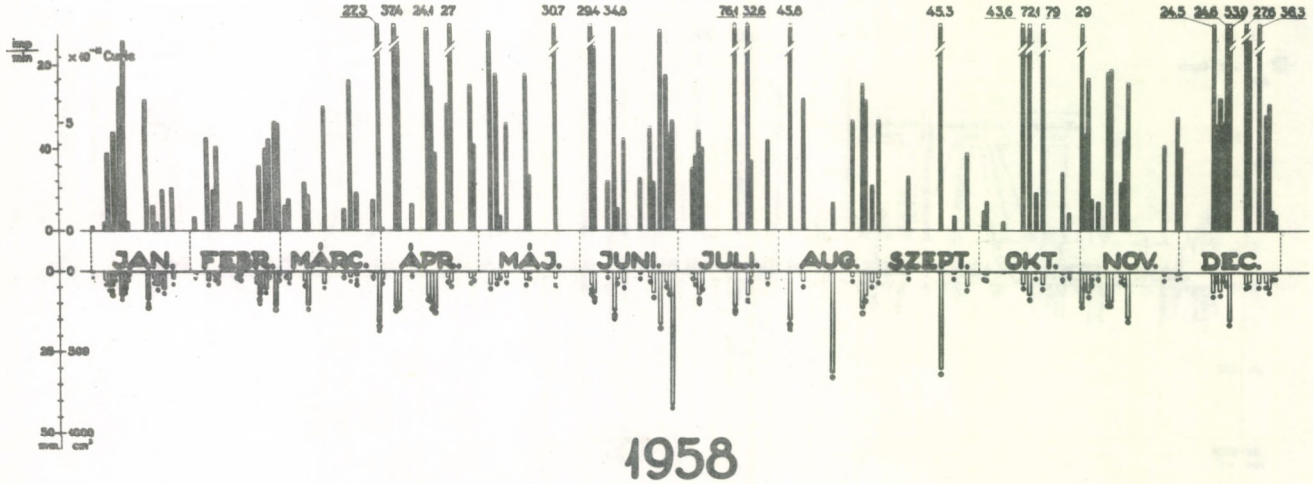

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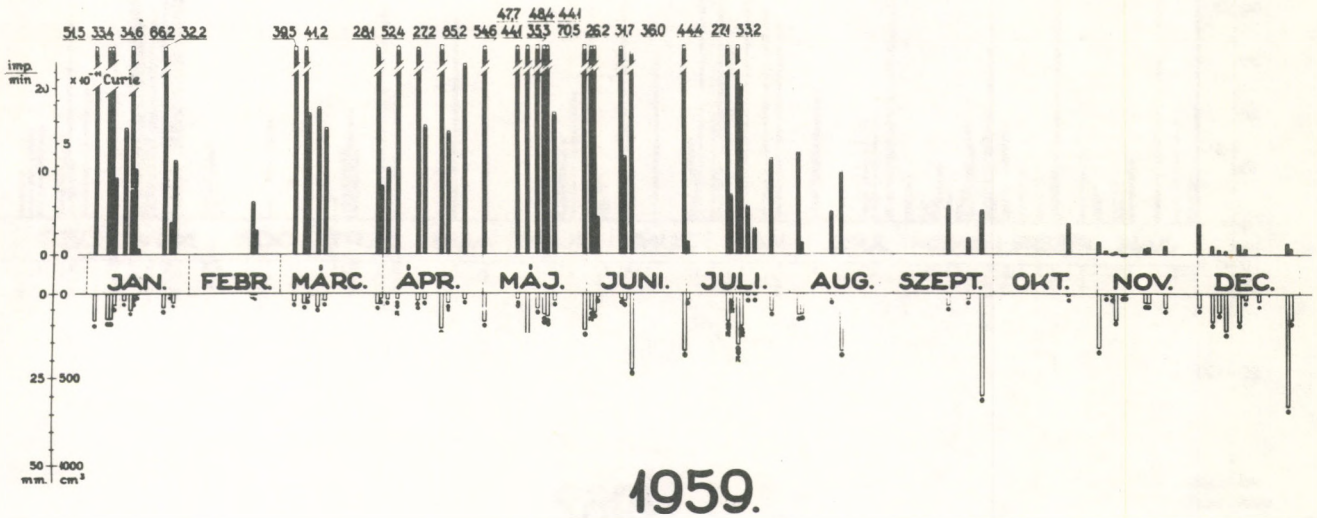

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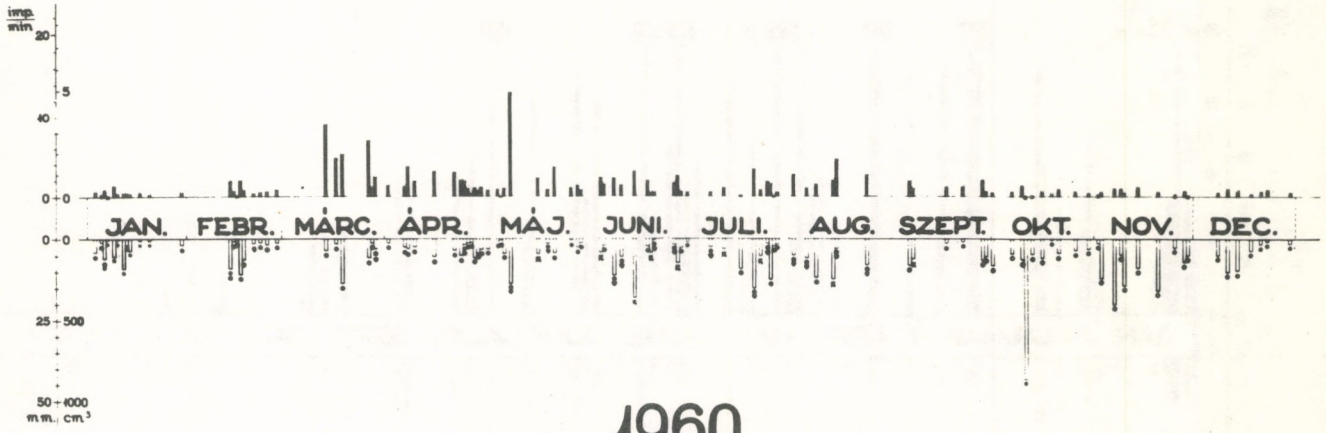

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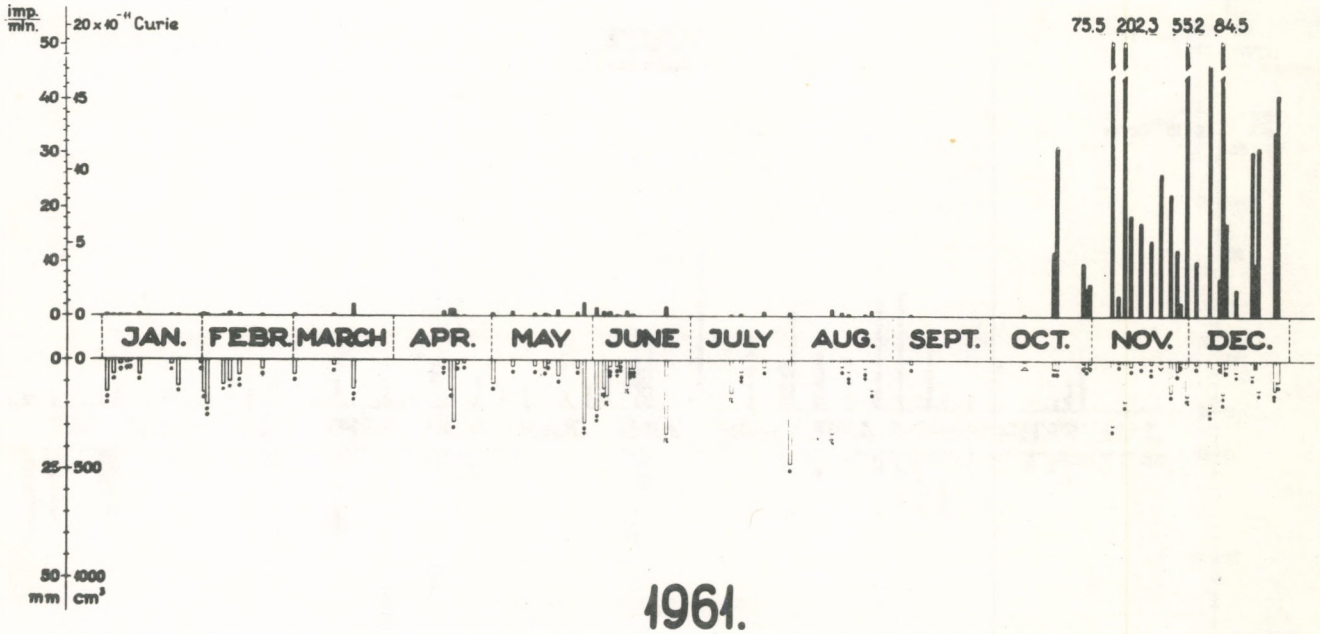

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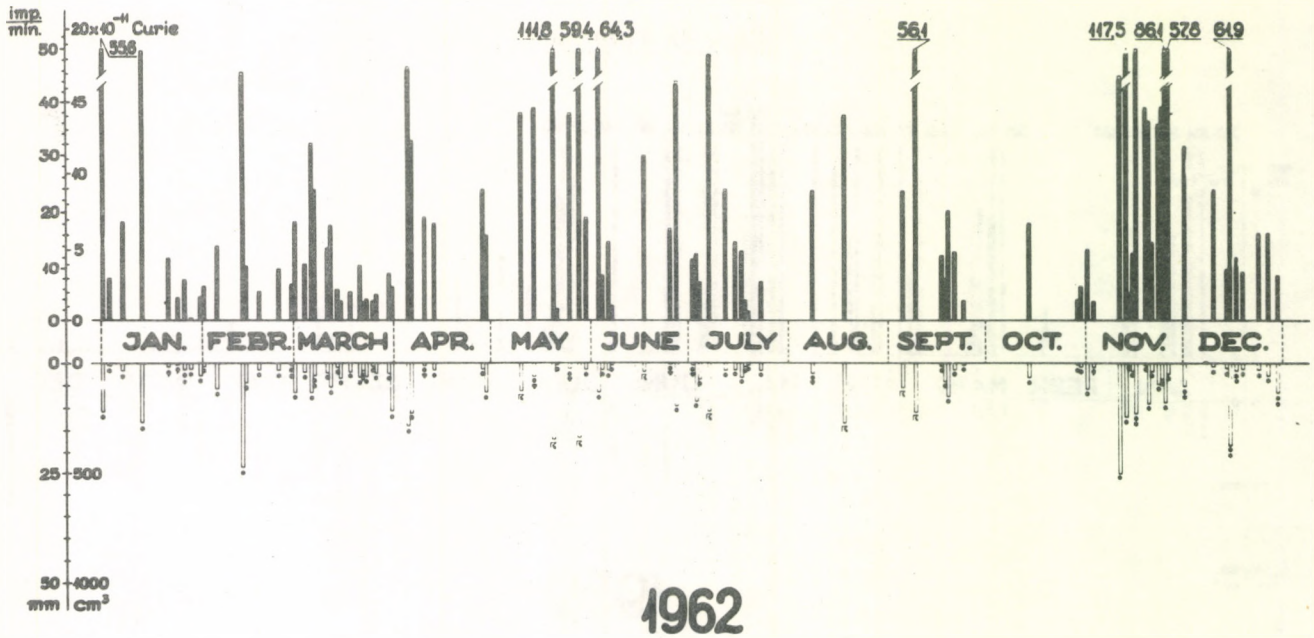

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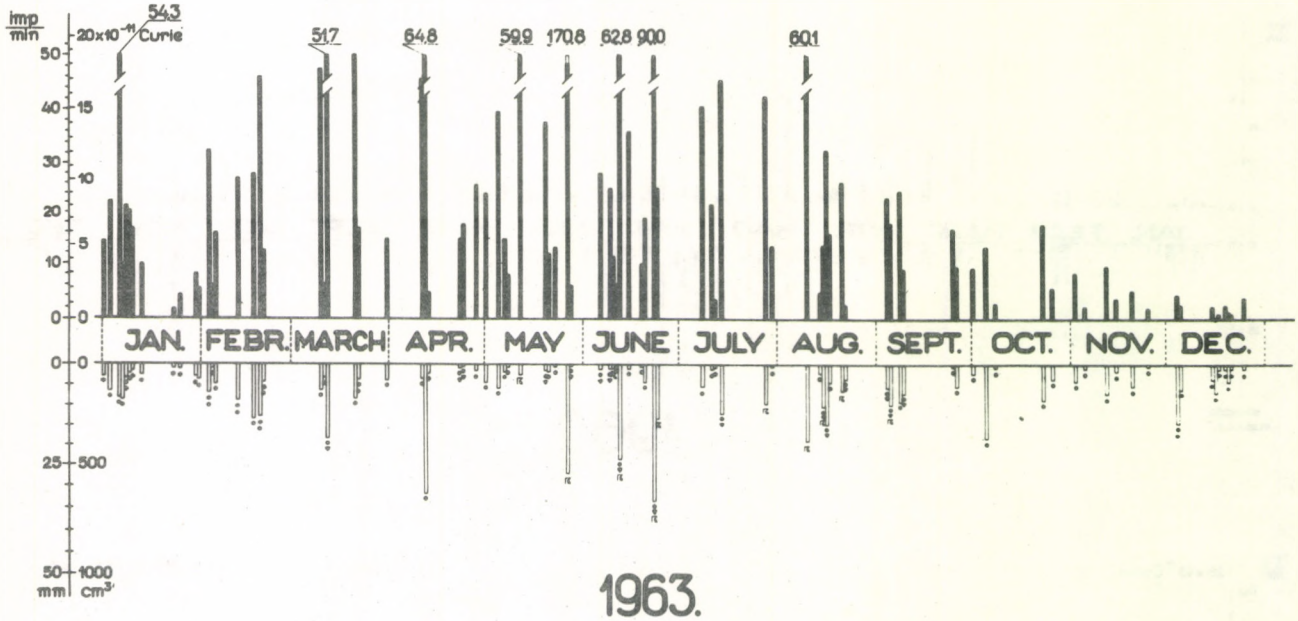

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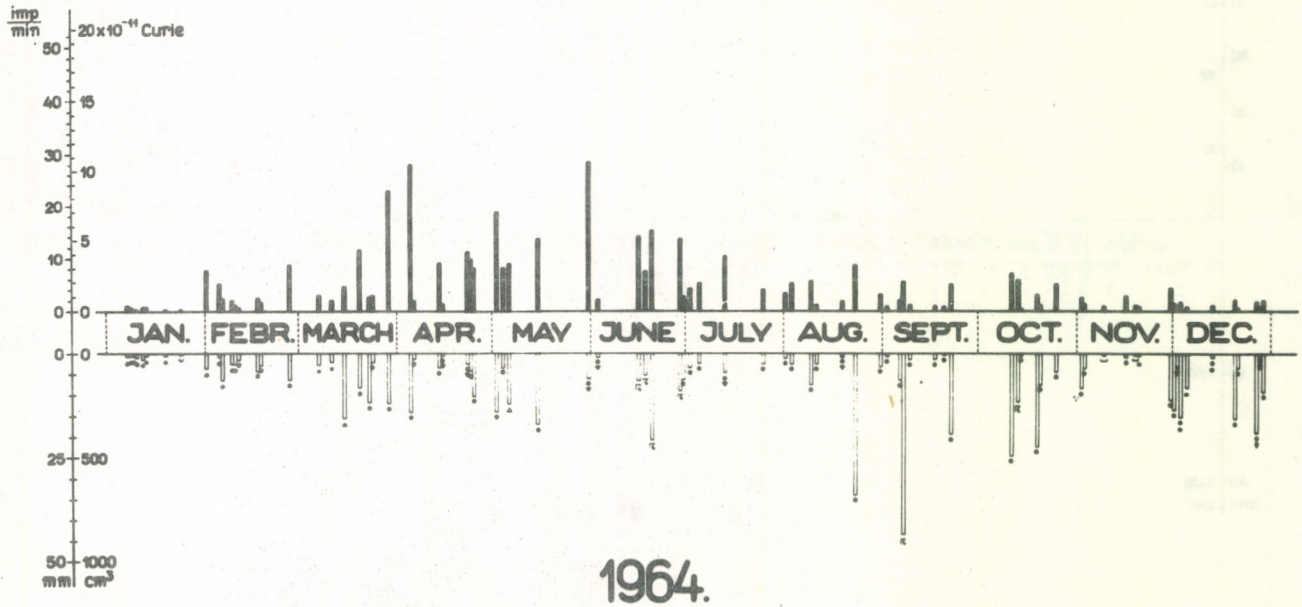

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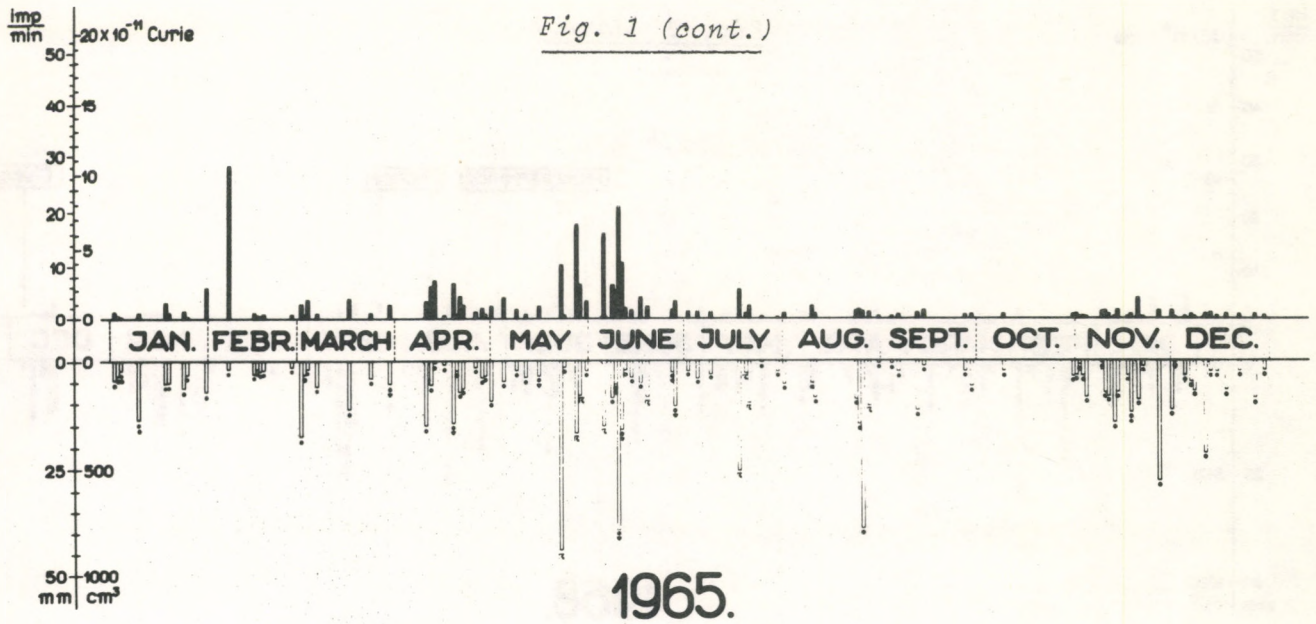

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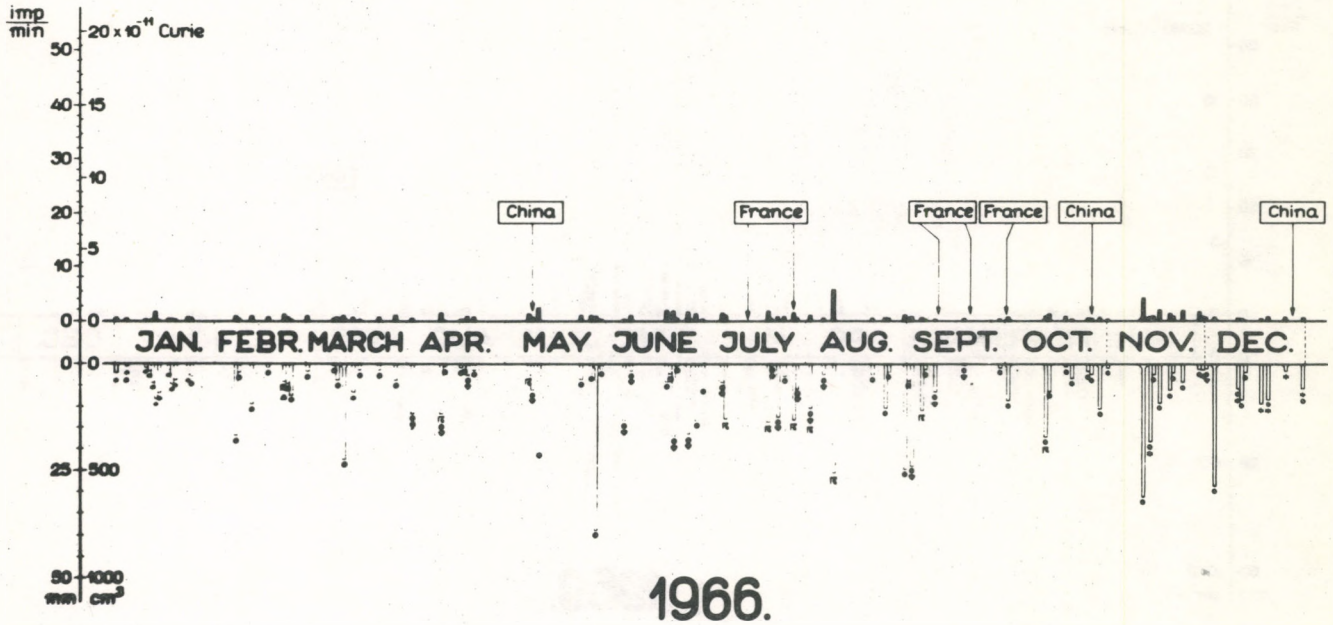

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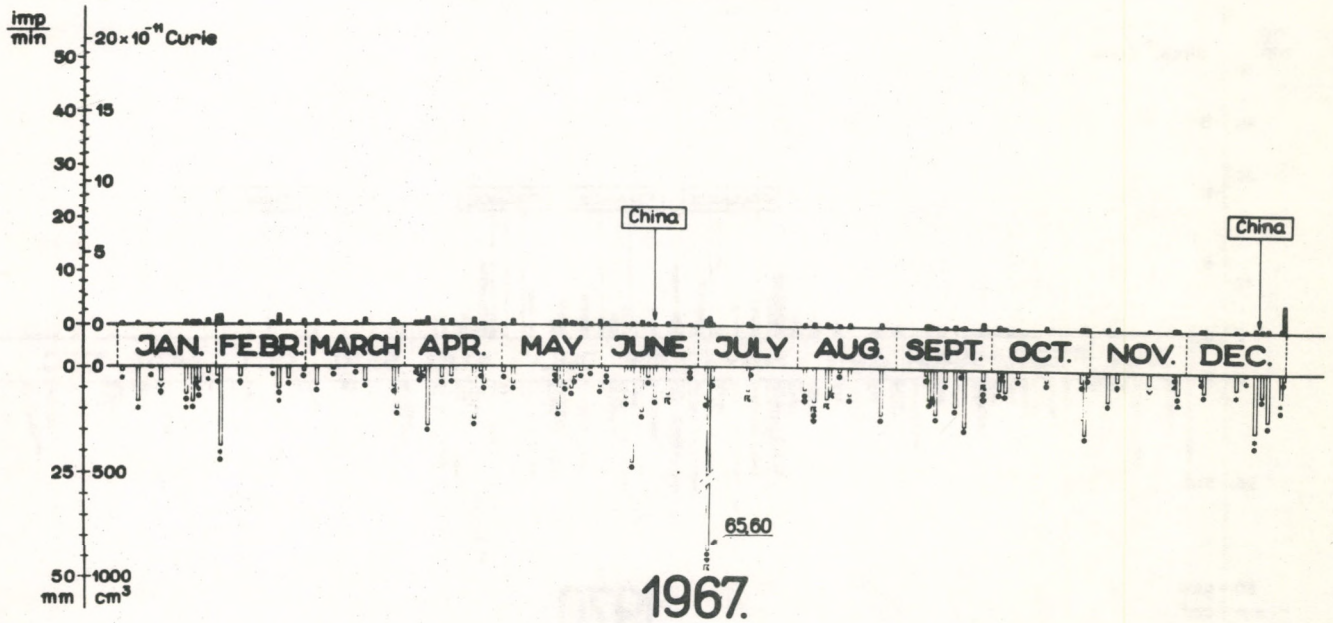

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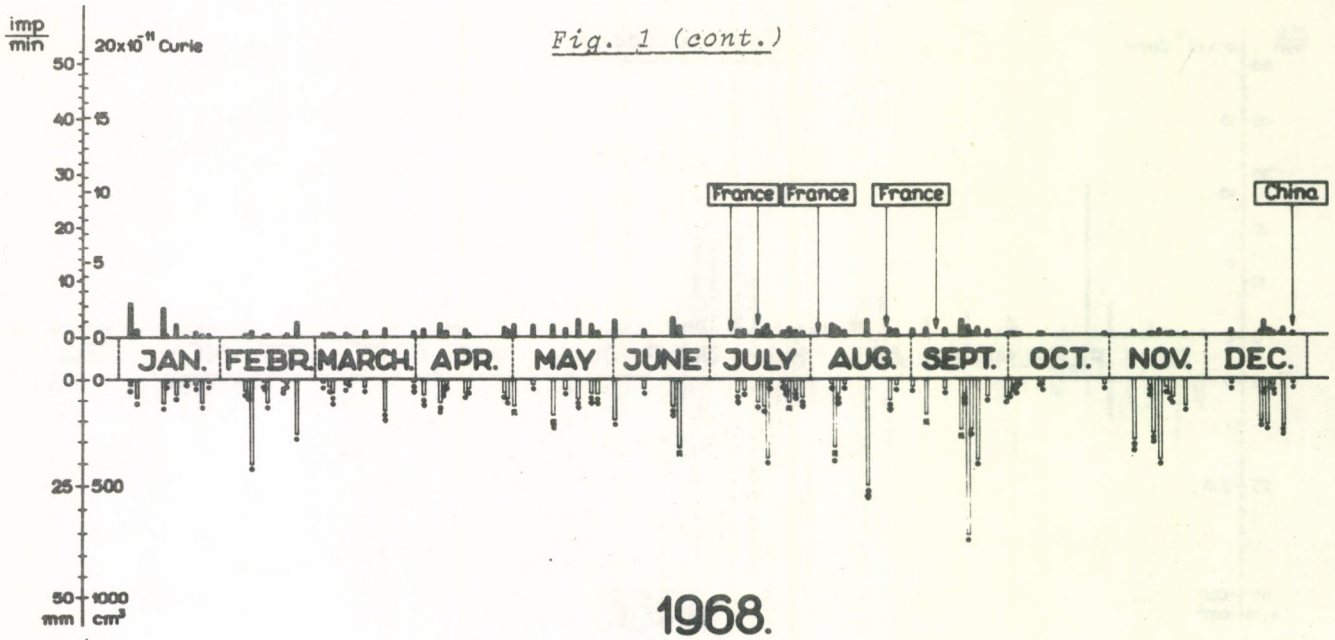

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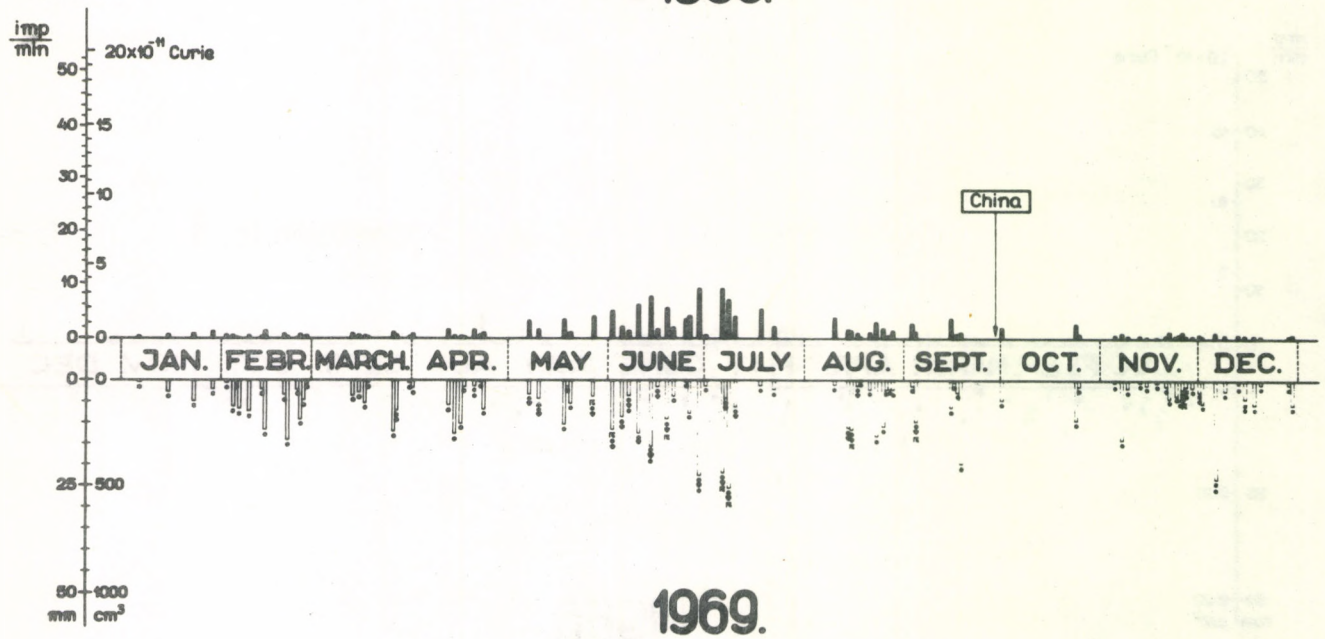

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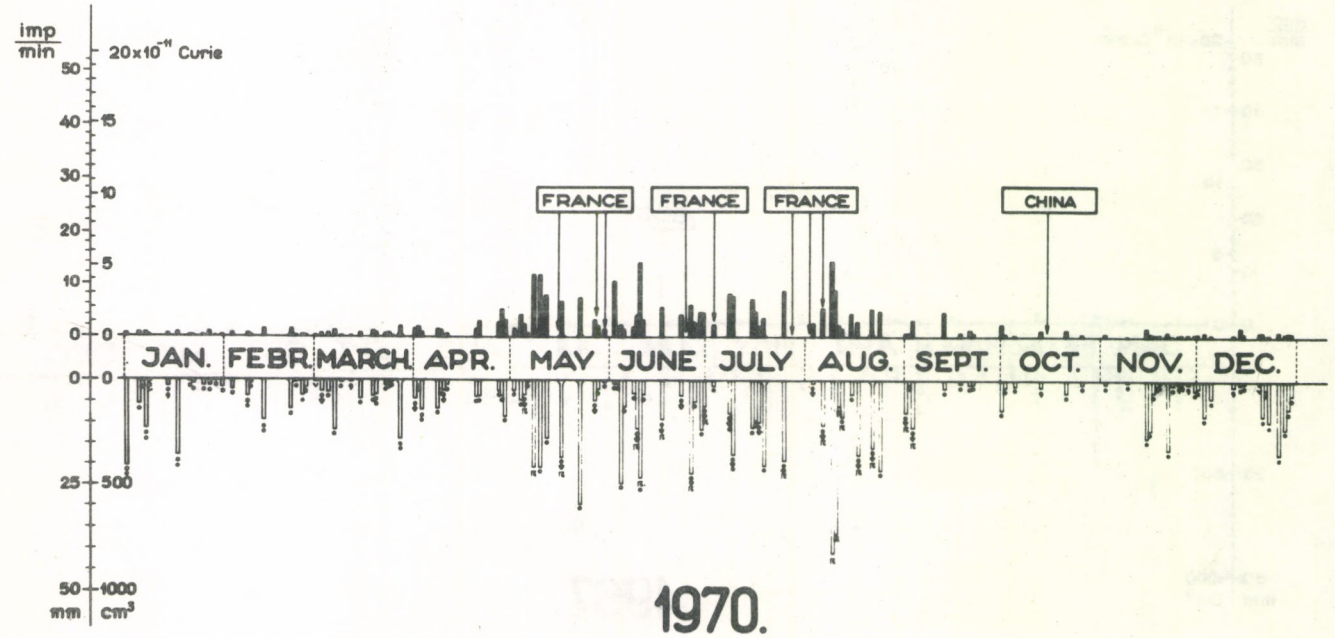

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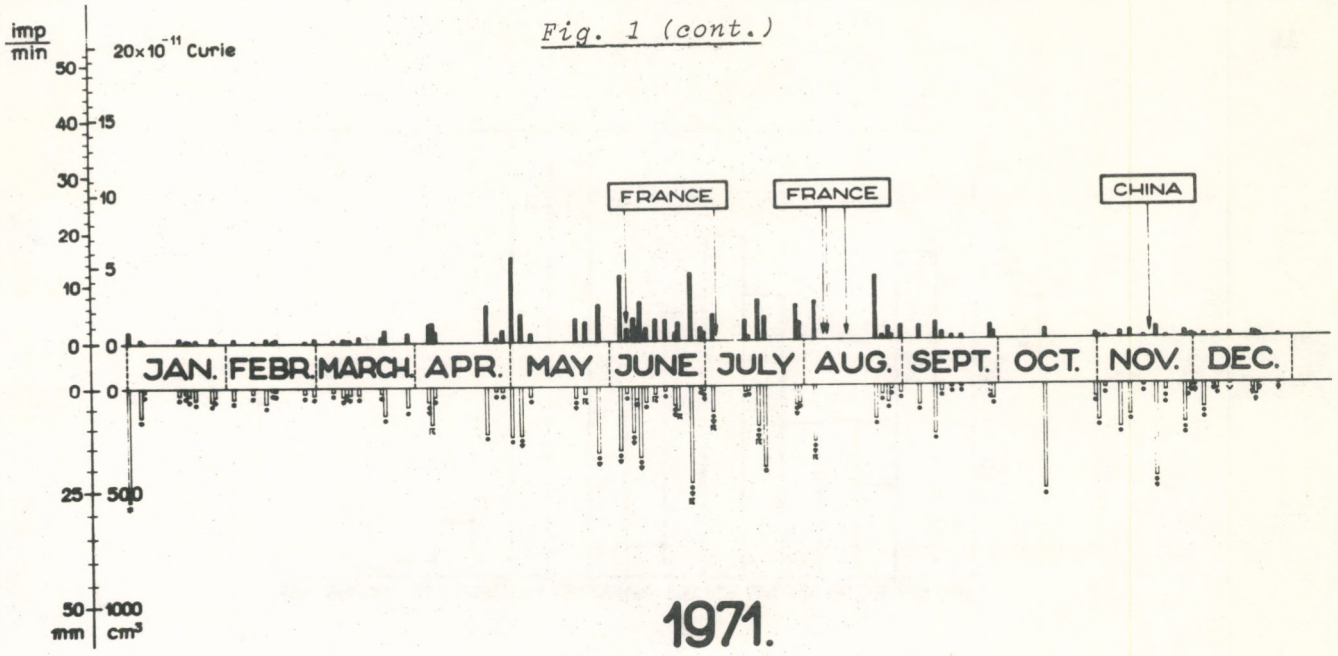

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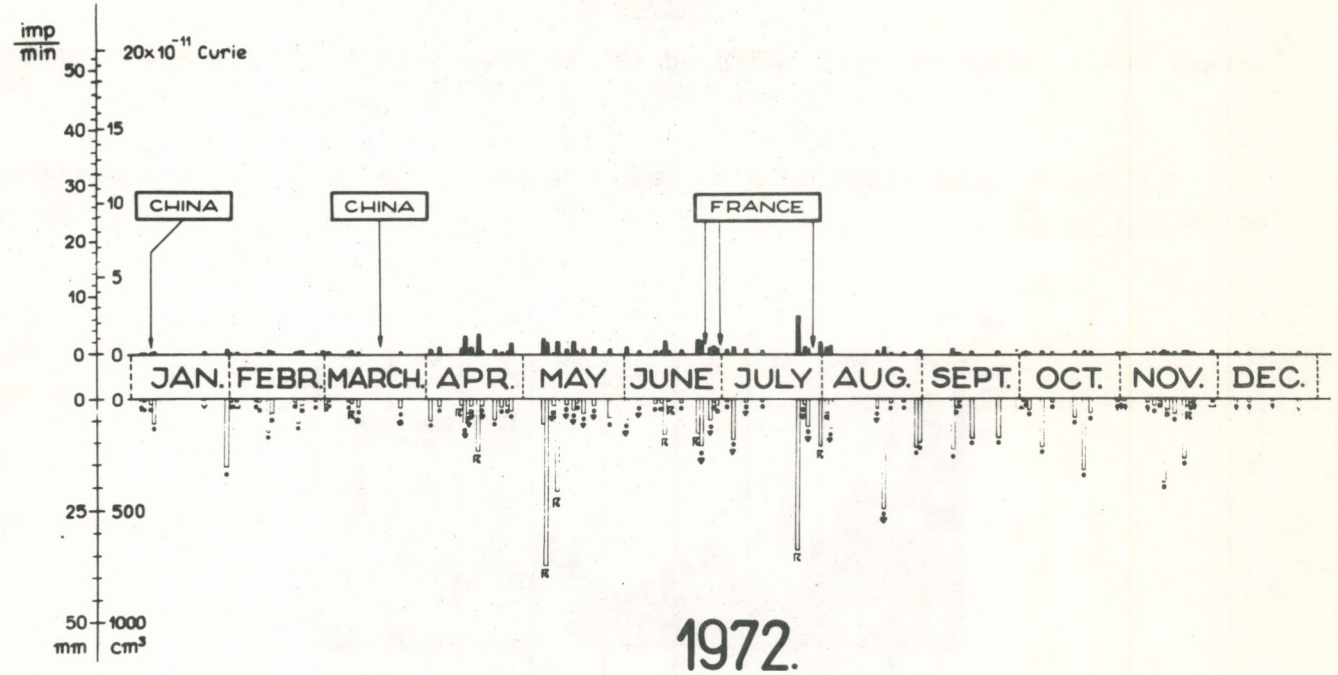

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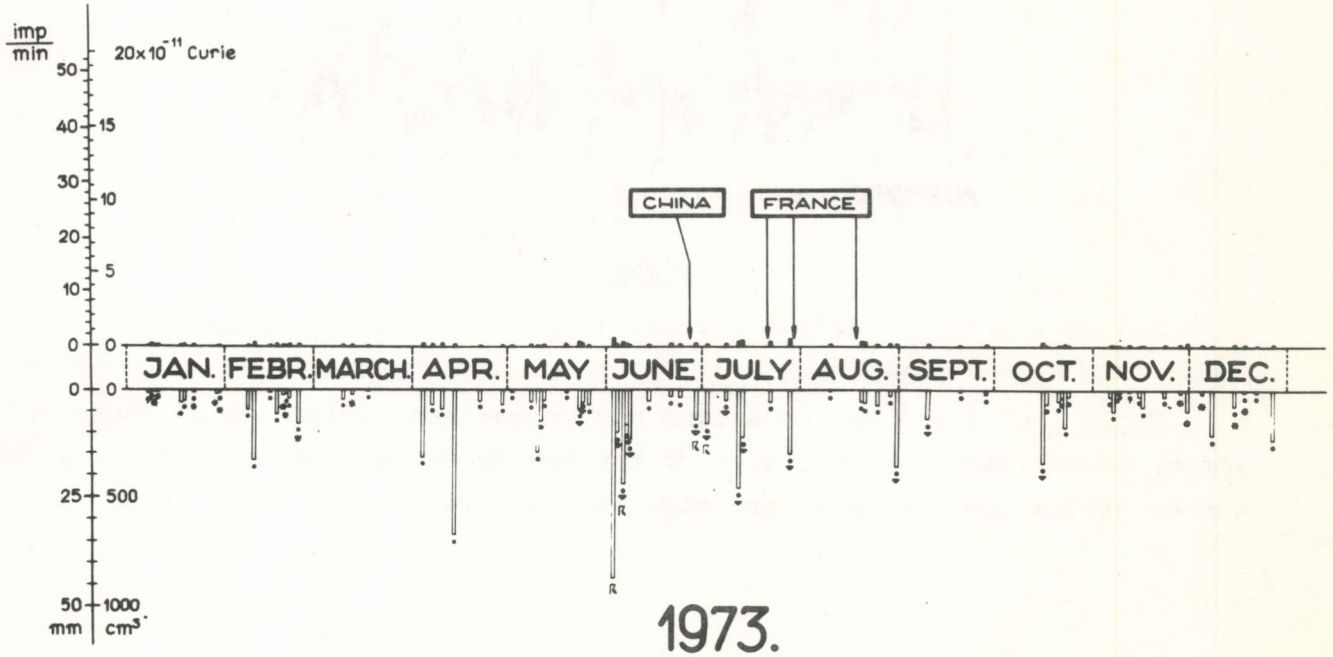

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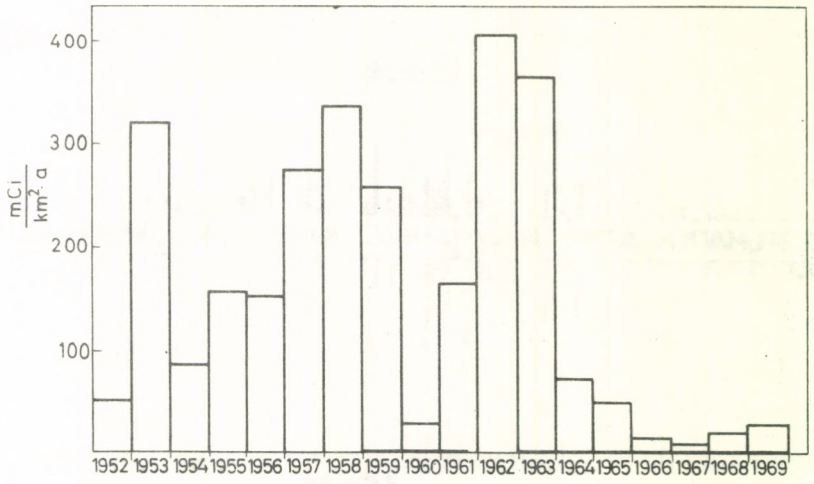

Figure 2

Annual sum of beta-activity measured in the precipitation in Debrecen, Hungary, between 1952 and 1969 [2]

The yearly summarised data between the years 1963 and 1971 are presented in Fig. 3 [3].

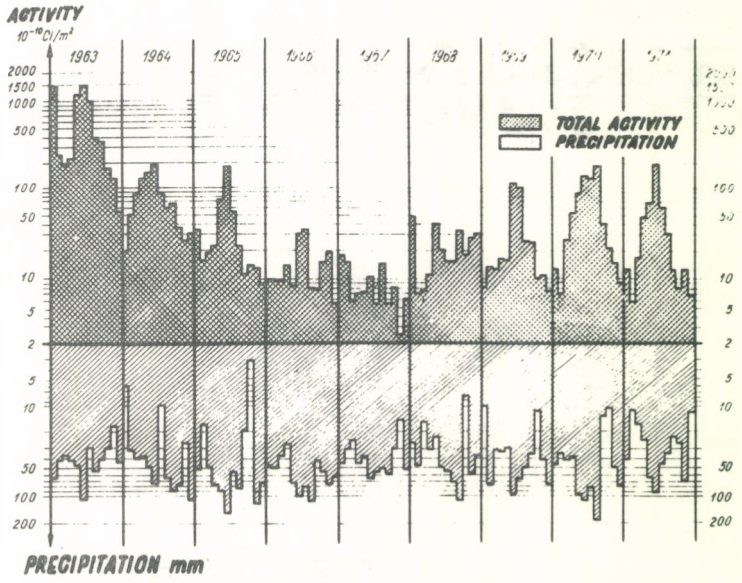

Figure 3

Radioactivity of fallout between 1963 and 1971 in Debrecen [3]

Table 1 contains the relations and summarised evaluation of these data, where, in addition to the values of the yearly summarised beta-activity, the number of the observed hot particles is also stated [3].

Year	Precipitation mm	Total beta activity, 10^{-10}Ci/m^2	Total activity, % of 1963 total activity	Number of hot particles	24-hr max. 10^{-10}Ci/m^2	24-hr max. % of 1963 max.
1963	578.1	6738	100	72	576.0	100
1964	613.9	921	13.6	26	49.0	8.5
1965	795.0	600	7.8	10	39.0	6.7
1966	780.7	176	2.6	3	12.0	2.2
1967	517.9	99	1.3	0	4.8	0.8
1968	549.4	285	4.2	0	9.9	1.7
1969	565.2	356	5.3	6	26.7	4.6
1970	788.3	712	10.6	21	32.1	5.6
1971	469.5	469	7.0	11	49.9	8.7

Table 1

Changes of the quantitative and qualitative characteristics of the activity of fall-out samples in Debrecen, between 1963 and 1971 [3]

The gross beta- and ^{90}Sr activity measured between 1963 and 1967 in Budapest are shown in Figs. 4 and 5 [4].

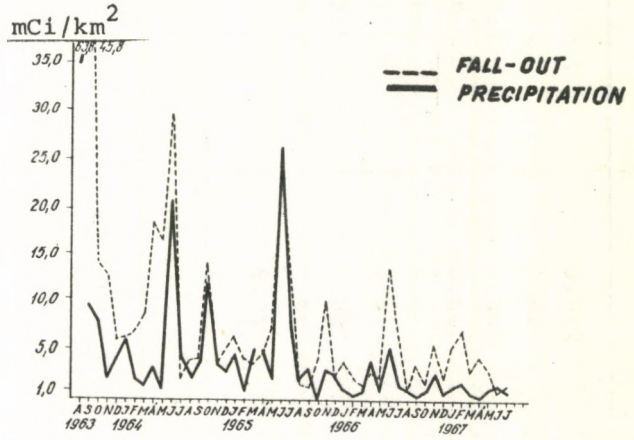

Figure 4

Gross beta-activity on the soil based on the measurements of fall-out and atmospheric precipitation in mCi/km² [4]. (The months are indicated by their initials)

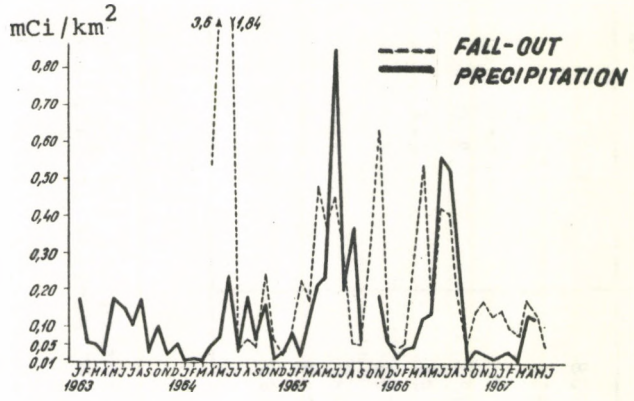

Figure 5

^{90}Sr activity on the soil based on the measurements of fall-out and atmospheric precipitation in mCi/km² [4]

The gamma-activity of fall-out was measured in one of the sampling station networks in Budapest. Table 2 contains the detailed monthly data measured at this station between 1964 and 1973 [5].

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

Table 2

Monthly averaged fall-out gamma-activity (in nCi/m^2) measured in the region of the Central Research Institute for Physics and the Institute of Isotopes between 1961 and 1973 [5]

The National Meteorological Service carried out the sample taking of the fall-out and the precipitation by the internationally developed measuring methods. Tables 3 and 4 demonstrate the total beta-activity of the fall-out and the precipitate relating to Budapest, Pécs and Szeged in the years from 1955 to 1973 [6, 23].

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	0,094	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

Table 3

Mean daily values of the total beta-activity of fall-out in mCi/km^2 [6,23]

Year	Activity, nCi/l		
	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

Table 4

Mean values of the total beta-activity of precipitation in nCi/l [6,23]

Fig. 6 [23] represents the annual mean value of precipitation activity data measured in Budapest as well as the maximum value measured in the actual 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.

From the evaluation of the results (see Table 1) it may be unanimously stated that the highest beta-activity has been measured in 1963 and simultaneously the highest number of hot particles has been observed. Following the atomic bomb ban treaty of 1963 in Moscow there was a rapid clearing of the atmosphere: in 1964 the fall-out values were already lower by one order of magnitude against the previous years. In 1965 the total radioactive material

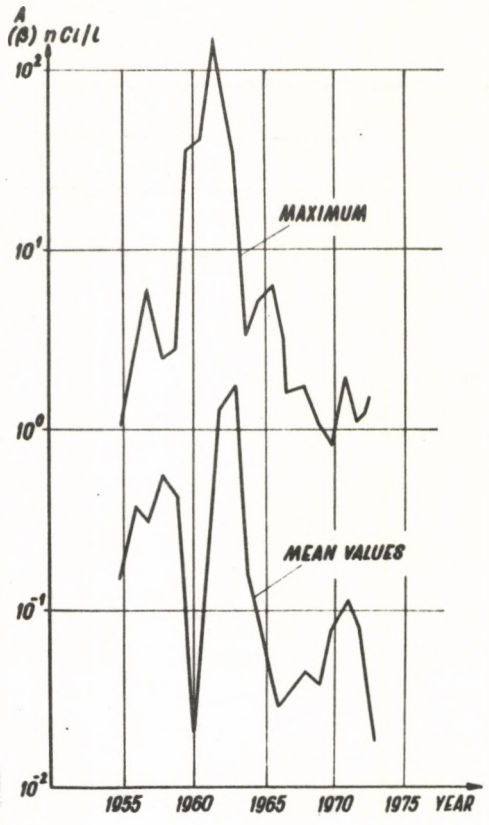

Figure 6

Comparison of the measured maximum radioactivity of precipitation with the annual mean value [23]

of the fall-out during the whole year was only slightly more than the maximum of a 24-hours fall-out observed in 1963.

The percentage of supplies is a satisfactory possibility for comparison. Taking the gross beta-activity of the fall-out in 1963 for 100%, there is a steady decrease during the following 4 years of the percentual values. In 1967 only 1.3% of the 1963 fall-out activity has been measured.

The data of 1968, however, cannot be fitted into the decreasing series. Presumably this is the consequence of renewed explosions carried out during this period which could be observed not only in the Hungarian data but in the literature too.

Aerosol Measurements

The systematic investigation of the radioactive aerosol contamination close to the surface of soil has been started within the scope of one of the sampling station networks in Budapest in 1961. The monthly data of the measurements between 1961 and 1973 are shown in Table 5 [5].

From another station in Budapest the changes of aerosol activity between 1963 and 1967 are demonstrated by Figs. 7 and 8 [4].

The decrease of the aerosol activity by about 80% in 1964 manifested itself in a similar characteristic way as in the fall-out.

At the National Meteorological Service the determination of the aerosol activity is carried out among internationally accepted conditions. Table 6 presents the data of aerosol total beta-activity measured in Budapest, Pécs and Szeged in the years from 1955 to 1972 [6, 23]. Fig. 9 shows the maximum and the mean values measured in Budapest [23].

Year / Month	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972	1973
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
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
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
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
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
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
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
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
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
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
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
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

Table 5

Monthly averaged radioactive concentration values (in pCi/m^2) measured in the atmospheric region of the Central Research Institute for Physics and the Institute of Isotopes between 1961 and 1973 [5]

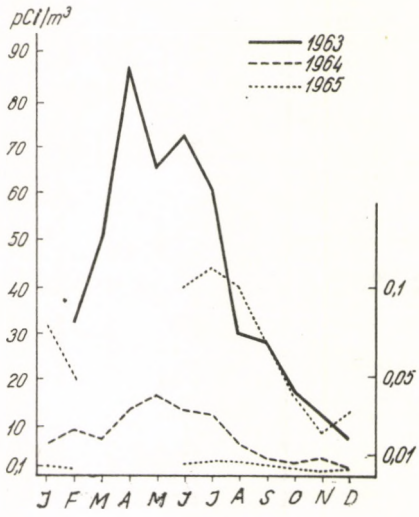

Figure 7

Monthly average of aerosol activity in pCi/m³ for 1963, 1964, 1965 [4]

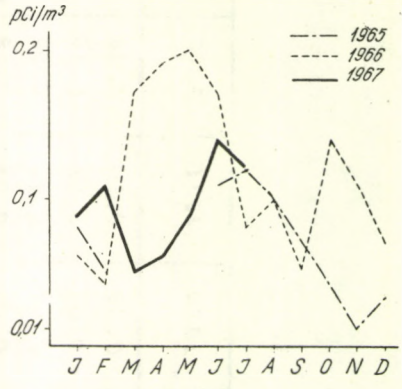

Figure 8

Monthly average of aerosol activity in pCi/m³ for 1965, 1966, 1967 [4]

Year	Activity, pCi/m³		
	Budapest	Pécs	Szeged
1955	1,25	-	-
1956	2,33	-	-
1957	1,81	-	-
1958	3,97	-	-
1959	4,50	-	-
1960	0,33	-	-
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

Table 6

Mean daily values of the total-beta activity of aerosol in pCi/m³ [6,23]

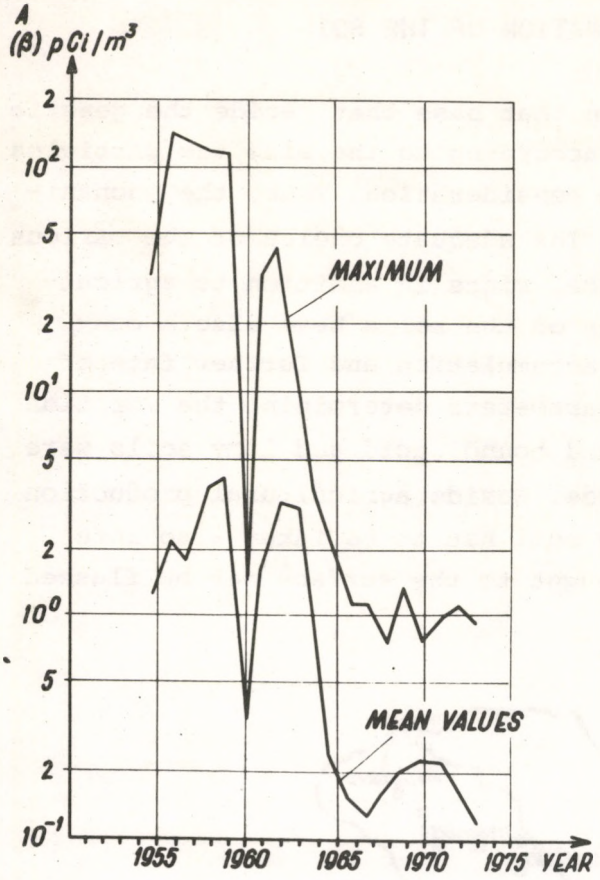

Figure 9

Comparison of the measured aerosol radioactivity with the annual mean value [23]

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

In the course of the fission process 36 noble gas isotopes are produced. These are partly stable, partly of short half-life, except ^{85}Kr of a half-life of 10.76 years, thus it may induce permanent radioactive contamination of the atmosphere. The measurements of ^{85}Kr activity are shown in Fig. 10 [7].

A continuous increase of the ^{85}Kr concentration in the air can be stated all over world. This fact is of special importance since the operation of the fuel reprocessing plants and so the establishment of new ones have a considerable influence on the ^{85}Kr concentration.

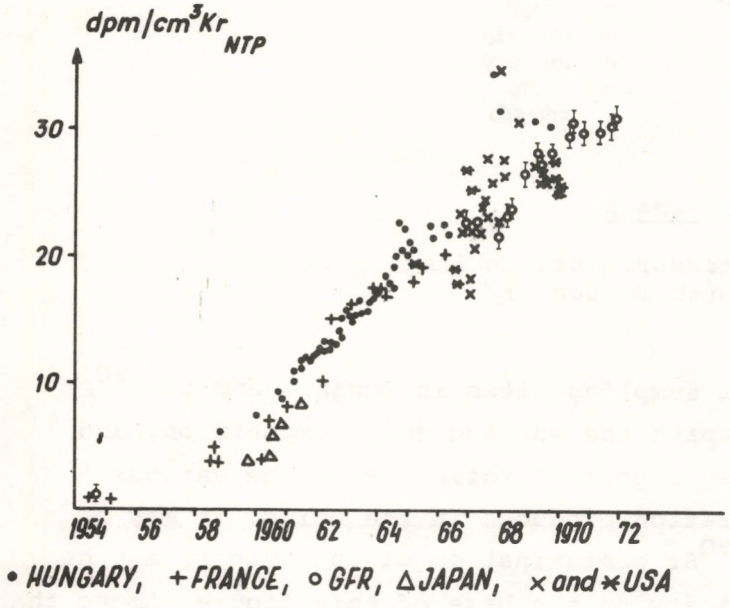

Figure 10

Increase of atmosphere ^{85}Kr concentration. The figure demonstrates the data measured in Debrecen between 1966 and 1971 with the published other data [7]

2. MEASUREMENT OF THE RADIOACTIVE CONTAMINATION OF THE SOIL

The sampling sites were determined on that base that beside the genetic classification of the soils the grouping according to the size the particles forming the soil has also been taken into consideration. Thus, the mechanical composition has been considered, too. The adequate choice of the various sampling sites is of outstanding importance, since in addition to agricultural techniques the adsorption properties of the soils have also a considerable influence on the distribution, accumulation and further fate of the contamination. Therefore, among the parameters determining the sorption of radioactive contamination, the sandy and bound, acid and limy soils were taken into consideration in the first place. Beside agricultural production and soil composition the structure of the soil has to be taken also into account, since the various substances brought to the surface may be flushed by streaming waters.

Figure 11

Sites of the soil activity measurements in Hungary and the ^{90}Sr activity values [8]

Fig. 11 demonstrates the soil sampling sites in Hungary and the ^{90}Sr activities measured there [8]. Despite the varying soil composition, and taking also into consideration the degree of influence of the various factors on the radioactive accumulation produced in the soils, it may be stated that the radioactive i.e. ^{90}Sr contamination of the country may be characterised with relative simplicity on the base of this figure. Among the

studied regions the majority belong into two groups, thus demonstrating a contamination of 100 - 150 and 150 - 200 pCi ^{90}Sr/g Ca content. Table 7 demonstrates the ^{90}Sr contamination results of the various soil samples measured, categorised according to the type of soil and cultivation [9]. The data of the table show the importance of the study of the soils cultivated by various means.

Soil	*Cultivation*	*Depth of sampling, cm*	*^{90}Sr activity, mCi/km^2*
Humus sand	*pasture*	*0 - 1*	*10.20*
		1 - 5	*0.30*
Humus sand	*ploughland*	*0 - 5*	*6.20*
		5 -10	*2.50*
		10-20	*3.40*
"Solonchak-solonyets"	*pasture*	*0 - 5*	*24.30*
"Solonchak"	*pasture*	*0 - 5*	*13.90*
"Solonchak-solonyets"	*pasture*	*0 - 5*	*20.60*
Flooded meadow land	*ploughland*	*0 - 5*	*8.05*
		5 -10	*6.75*
Meadow soil	*pasture*	*0 - 5*	*11.10*
Meadow soil	*ploughland*	*0 - 5*	*1.10*
"Solonyets" meadow soil	*pasture*	*0 - 5*	*11.90*
Deep meadow "solonyets"	*ploughland*	*0 - 5*	*15.30*
Deep meadow "solonyets"	*ricefield*	*0 - 5*	*4.30*

Table 7

^{90}Sr pollution of soils according to the type and cultivation [9]

Fig. 12 demonstrates the ^{90}Sr contamination of the soils and its yearly increase between 1958 and 1968 [10].

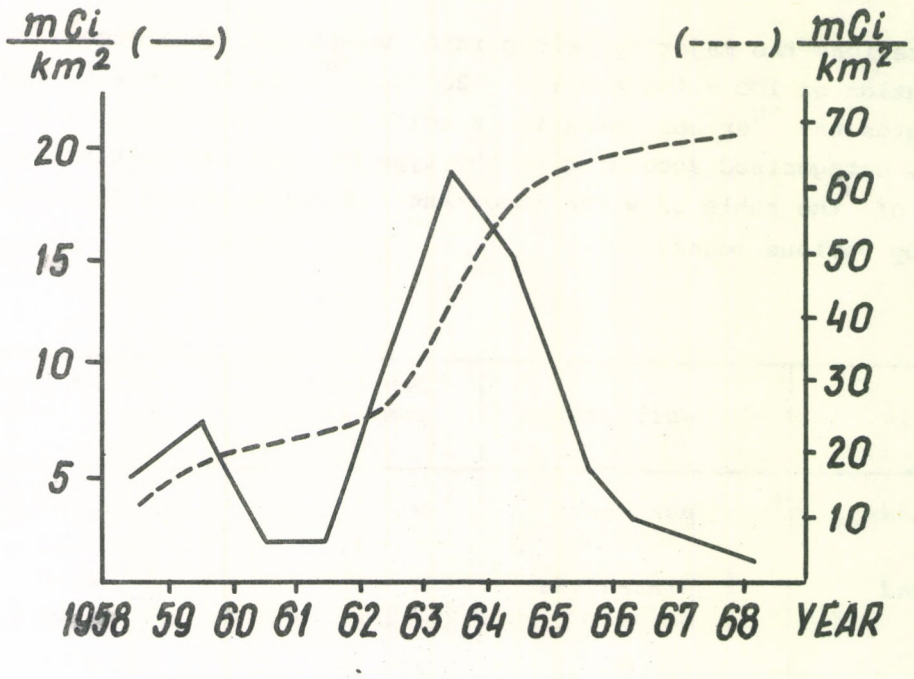

Figure 12

^{90}Sr contamination of the soil (——) and its yearly increase (----) [10]

Fig. 13 demonstrates that a correlation of the ^{90}Sr contamination may be stated between the soil and vegetal samples originating from the same regions /8/.

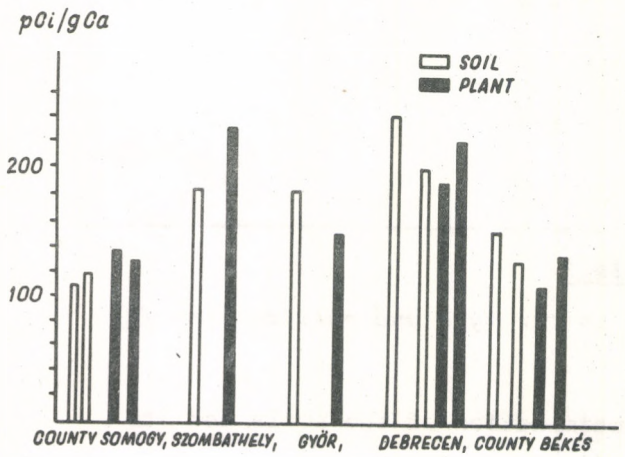

Figure 13

^{90}Sr contamination of soil and vegetal samples orginating from identical regions [8]

In addition to the ^{90}Sr contamination studies on the determination of the ^{137}Cs contamination of the same type soils has also been carried out with due emphasis. Fig. 14 demonstrates the ^{137}Cs contamination and the yearly accumulation in the soils between 1961 and 1968 [10].

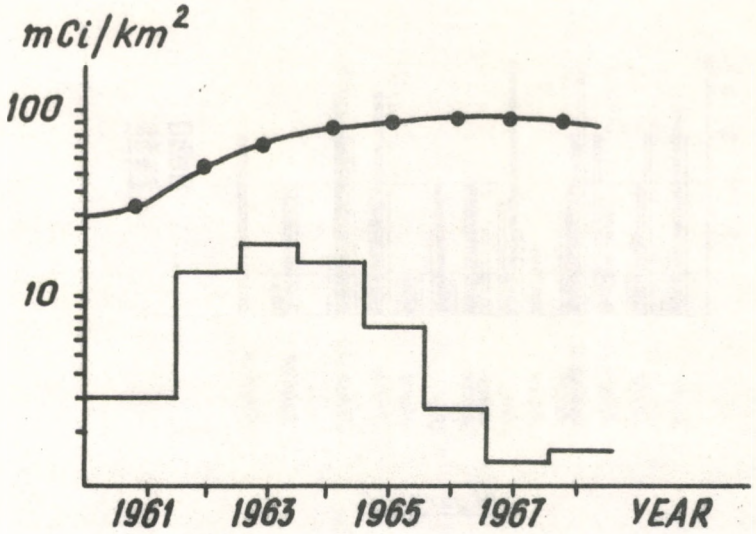

Figure 14

^{137}Cs contamination of the soil and its yearly accumulation [10]

To the environmental radioactivity data, especially to the soil contamination, it should be emphasised that the highly radiotoxic and long lived ^{90}Sr and ^{137}Cs isotopes were measured systematically by utilising a large number of samples.

Based upon the soils of various composition those soilphysical and chemical factors could also be analysed, which, owing to their chemical properties similar to those of the actual radioactive elements (e.g. high Ca or high K concentration), in consequence to the natural composition would influence also the expected contamination of the plants cultivated in the actual region.

The data prove that artificial radioactive contamination is concentrated chiefly in the surface layers of the soil.

3. MEASUREMENT OF THE RADIOACTIVE POLLUTION OF SURFACE WATERS

The radioactive pollution of the rivers and lakes of Hungary is regularly controlled since the middle fifties. The majority of the rivers comes 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.

Figs. 15 and 16 show the total beta-activity of the Danube, the Tisza and their tributaries between 1963 and 1966 [11].

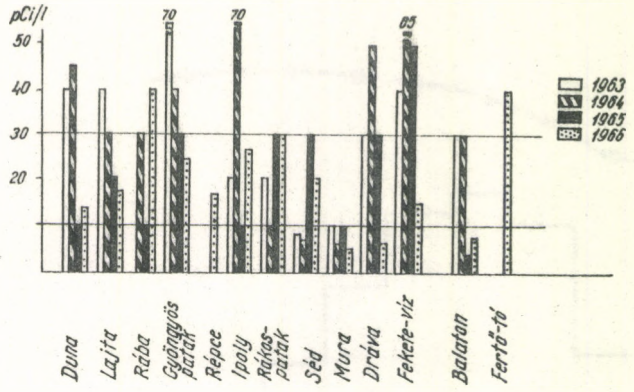

Figure 15

Total beta-activity of the Danube and its tributaries in the years 1963-1966 [11]

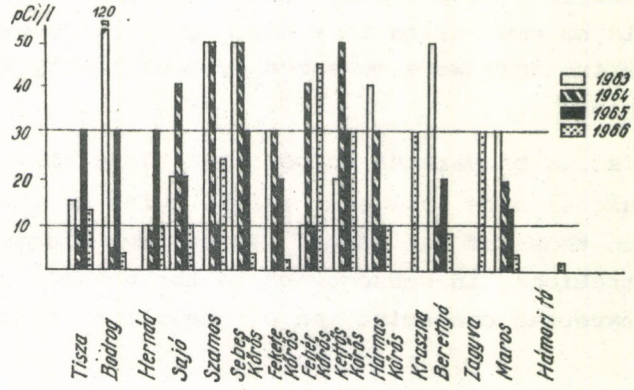

Figure 16

Total beta-activity of the Tisza and its tributaries in the years 1963-1966 [11]

From another series of measurements the contamination of some of the larger rivers of Hungary and the Lake Balaton are presented between 1965 and 1971 in Table 8 [12, 13].

The total beta-activity of the Danube and its seasonal distribution is demonstrated by Fig. 17 [12].

Year	Danube	Rába	Dráva	Tisza	Sajó	Balaton
1965	0.13	0.36	0.16	0.14	0.40	0.11
1966	0.09	0.29	0.14	0.15	0.34	0.09
1967	0.09	0.30	0.13	0.16	0.50	0.09
1968	0.03	0.44	0.13	0.14	0.40	0.07
1969	0.04	0.37	0.11	0.14	0.13	0.06
1970	0.09	0.23	0.15	0.15	0.23	0.15
1971	0.08	0.29	0.17	0.05	0.18	0.16
1972	0.08	-	-	-	-	-
1973	0.10	-	-	-	-	-

Table 8

The average gross beta-activity of surface waters in Hungary in x10^{-7} µCi/ml [12,13]

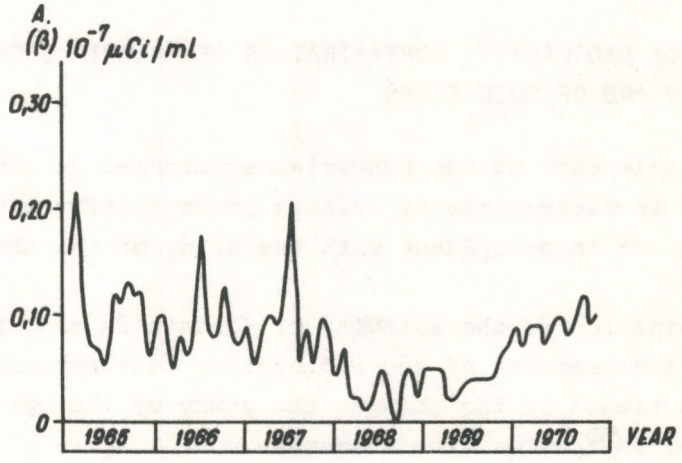

Figure 17

Total beta-activity of the Danube river [12]

The activity of the metal ion fraction and of ^{137}Cs in the Danube at Budapest between 1969 and 1973 are shown in Table 9 based on Refs. [13,24].

Year	^{137}Cs activity, pCi/l		Metal ion fraction activity, pCi/l	
	Average	Max.	Average	Max.
1969	1.09	4.0	0.55	2.0
1970	1.66	3.0	1.85	5.0
1971	1.68	4.0	1.83	5.0
1972	0.90	4.0	0.87	4.0
1973	1.24	9.0	0.86	2.0

Table 9

Activity of ^{137}Cs and metal ion fraction in the Danube at Budapest in the years 1969-1973 based on Refs. [13, 24]

Around the year 1963 the total beta-activity of the surface waters was rather high, then decreased gradually. Between 1965 and 1970 it was in general - expect the rivers Rába and Sajó of 10 to 15 pCi/l activity. The average activity of the metal ion fraction and ^{137}Cs changed in the years 1969 to 1973 between 0,55 pCi/l and 1,85 pCi/l.

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

A considerable part of the Hungarian environmental monitoring studies consists of the investigations of various products of vegetal and animal origin, carried out in accordance with the study of the atmosphere, soil and water.

The determination of the activity of foodstuffs also 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 plants supply excellent samples for the determination of the level of environmental contamination as well as of its changes. The radioactive elements falling directly on the foliage or entering the plants through the soil may be determined simply and exactly. The contamination studies covering a longer period allow to acquire information concerning prolonged time intervals by utilising vegetal samples of various vegetation periods. However, the estimation of the risk of contamination includes also the knowledge of

the discrimination represented by the animal metabolism, therefore it seems advisable to determine the contamination of vegetal and animal organisms in samples taken at the same time from identical regions.

The choice of the sampling sites was performed on the base of the territorial properties (see Figs. 18 and 19) [14]. Their uniform distribution makes possible the adequate information from the whole country.

Figure 18

Sampling sites for vegetables and milk in Hungary [14]

Figure 19

Sampling sites for bone and others in Hungary [14]

Supplementing the data it has to be emphasised 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 (common sorrel) were studied for statistical evaluation. These vegetables represent a regularly consumed group of products, and they supply reliable information concerning the quantity of materials taken up either from the fall-out or from the soil (with special regard to metal ions).

The data of the systematic investigations were evaluated from three chief sampling districts: I. Transdanubia; II. the region between the rivers Danube and Tisza; III. Trans-Tisza region.

The yearly averaged values of the data concerning spinach, lettuce and oxalis, utilised also for statistical evaluation are shown in Tables 10-12 [14].

Time of sampling	*Area of sampling*		
	I. Transdanubia	*II. 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*

Table 10

Activity of metal ion fraction indicating the radioactive contamination of spinach, pCi/g dry substance [14]

Time of sampling	Area of sampling		
	I. Transdanubia	II. Region between Danube and Tisza	III. Trans-Tisza 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

Table 11

Activity of metal ion fraction indicating the radioactive contamination of lettuce pCi/g dry substance [14]

Time of sampling	Area of sampling		
	I. Transdanubia	II. Region between Danube and Tisza	III. Trans-Tisza 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

Table 12

Activity of metal ion fraction indicating the radioactive contamination of oxalis, pCi/g dry substance [14]

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

The results of the variance analysis may be summarised as follows:

(1) From the spring of 1962 to the end of the year 1964 the contamination of each sample was significantly higher than in the previous, and during the following years;

(2) Based upon the analysis according to the regional distribution, significant differences between the various sampling sites could not be observed;

(3) Neither the specific control examinations nor the results of the regular measurements demonstrated regions of such consequently higher contamination, which should be qualified for particularly contaminated or are to be submitted to increased control from the point of view of fall-out or production.

Measurement of Fodder and Crops

The results of fodder examinations performed from 1965 to 1973 are demonstrated in Table 13 [15].

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	-	-	3.9	-	-

Table 13

Radioactive contamination of fodder in the years 1965-1973. Activity of metal ion fraction in pCi/g dry substance [15]

The yearly averages of both hay and silage are rather uniform and their activity is within the range of the same order of magnitude as that of the indicator vegetables i.e. spinach, lettuce, oxalis (see Tables 10-12).

The characteristics of the crops and the products prepared of them are summarised in Table 14 (based on Ref. [16]) and Table 15 [15].

Sample	Ash content, %	Total beta activity	^{40}K activity	Residual beta activity
Wheat				
Average	1.7	4.24	2.92	1.32
Maximum	2.4	7.15	4.68	2.79
Minimum	0.8	1.33	1.21	0.09
Rye				
Average	1.9	5.40	3.86	1.54
Maximum	2.4	6.15	2.90	2.89
Minimum	1.7	4.33	4.27	0.06
Rice				
Average	1.25	2.23	0.66	1.56
Maximum	1.3	3.46	0.75	2.71
Minimum	1.2	0.99	0.58	0.41

Table 14

Radioactive contamination of crops in the years 1965-1968 in pCi/g substance (Based on Ref. [16])

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
Wheat flour	1971	1.23	0.04	1.22
	1972	1.8	0.1	1.6
Wheat bran	1971	11.23	0.36	9.83
	1972	10.5	0.3	9.1
Rye	1971	5.26	0.16	4.69
	1972	-	-	-
Rye flour	1971	2.22	0.9	2.1
	1972	1.9	0.1	1.7
Rye bran	1971	11.4	0.6	8.8
	1972	-	-	-
Rice	1971	1.4	0.08	1.22
	1972	1.7	-	1.2

Table 15

Radioactive contamination of crops and their products in the years 1971-1972 in pCi/g dry substance [15]

Measurement of Consumers' Goods

The examination serves as a control of the radioactivity of tobacco and the products arriving from foreign countries to Hungary (see Tables 16 and 17) [15, 16].

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

Table 16

Radioactive contamination of tobacco, in the years 1971-1973 in pCi/g dry substance [15]

Sample	Ash content, %	Total beta activity	^{40}K activity	Residual beta-activity
Tea				
Average	6.1	17.44	10.35	7.08
Maximum	6.8	21.76	16.36	11.20
Minimum	5.6	14.00	8.05	4.36
Coffee (green)				
Average	2.7	13.17	8.94	4.23
Maximum	4.5	18.99	11.73	7.26
Minimum	1.8	7.27	5.85	1.40
Cocoa bean				
Average	3.6	12.27	10.10	2.17
Maximum	3.9	13.83	12.06	2.94
Minimum	3.3	11.22	8.80	1.58
Cocoa powder	6.0	16.00	12.57	3.43

Table 17

Radioactive contamination of consumers' goods from 1969 in pCi/g [16]

Measurement of Milk and Dairy Products

The sampling sites of the milk are shown in Fig. 20 [17].

Figure 20

Origin of milk and dairy samples [17]

Table 18 (based on Refs. [17,18]) and Table 19 [19] contain the results of the representative measurements of dairy products.

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

Measurement of Fish

The measurement of fish samples of various origin supplies information concerning the degree of pollution occuring among river- and lake-keeping circumstances see Table 20 [15]. It appears clearly that the activity of the metal ion fraction characteristic to the artificial radioactivity is higher in the fish-bones than in the muscles.

A difference can be also stated between the contamination of fish originating from rivers and from lakes, namely the contamination of the fish samples taken from the lakes is higher. There is also a difference among the various fish species, but no more differences than one order of magnitude occured.

Measurement of Meat and Meat Products

The results of a representative measurement performed in 1965 are summarised in Table 21 [16].

Product and season	Ash content, %	Total activity, pCi/100 g	Activity calculated from K, pCi/100 g	Residual beta-activity pCi/100 g	Oxalat precipitation activity pCi/100 g	$^{90}Sr/gCa$ pCi/g
Milk,						
winter	0.66	125.9	114.1	11.8	6.5	4.1
summer	0.66	128.8	112.1	17.7	4.8	14.0
autumn	0.70	138.3	119.0	19.3	7.3	13.5
Cheese						
winter	4.34	134.3	101.0	33.3	21.3	5.4
summer	4.13	163.2	111.2	52.0	37.8	15.0
autumn	3.86	133.8	71.3	62.5	32.1	12.0
Curd						
winter	1.50	149.5	111.9	37.6	12.9	26.0
summer	1.55	148.1	99.5	48.6	11.1	43.0
autumn	1.53	160.0	106.6	54.3	11.6	64.0
Milk powder						
winter	7.03	1243.0	1136.3	106.7	69.8	6.0
summer	5.75	1136.5	998.5	138.0	40.4	16.0
autumn	5.62	1189.3	1014.4	174.9	40.8	19.0
Cream						
winter	0.30	62.2	53.4	8.8	4.4	59.0
summer	0.41	78.4	67.7	10.7	1.9	228.0
autumn	0.44	98.3	77.3	21.0	3.8	214.0

Table 18

Activity of milk and dairy products in the years 1961-1962. (Representative measurements based on Refs. [17,18])

Year	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

Table 19

Activity of the metal ion fraction of milk in the years 1960-1973 in pCi/100 g milk [19]

Sample (year)	Bone			Muscles		
	Total activity	Activity of metal ion fraction	^{40}K activity	Total activity	Activity of metal ion fraction	^{40}K activity
River fish /1971/						
average	5.0	2.2	-	6.0	0.2	8.0
maximum	11.0	7.8	-	17.2	1.4	15.1
minimum	0.0	0.0	-	1.9	0.0	1.6
/1972/						
average	6.0	2.0	2.4	9.9	0.3	9.0
maximum	7.0	3.1	3.8	11.8	0.5	11.1
minimum	3.8	0.4	1.4	7.3	0.2	7.3
Lake fish /1971/						
average	7.1	5.3	-	4.9	0.04	6.1
maximum	14.6	14.6	-	10.2	2.9	9.1
minimum	0.0	0.0	-	3.0	0.0	2.8
/1972/						
average	10.1	0.3	8.4	11.6	16.5	1.7
maximum	16.0	0.7	12.6	29.3	20.2	3.9
minimum	2.8	0.1	2.6	3.9	1.1	0.2
Various fish samples /1973/						
average	9.1	5.4	-	10.6	0.3	8.8

Table 20

Radioactive contamination of fish in the years 1971-1973 in pCi/g dry substance [15]

Sample	Ash content, %	Total activity	^{40}K activity	Residual beta-activity
Meats				
Pork	1.0	2.75	2.36	0.39
Beef	1.2	2.99	2.44	0.55
Duck	0.7	1.55	1.21	0.34
Meat products				
Sausage	2.6	1.81	1.31	0.50
Various	2.7	2.60	1.77	0.83

Table 21

Radioactive contamination of meat and meat products in 1965 in pCi/g [16]

Radioactive Contamination of Animal Bones

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

The results of the investigation of horned cattle bones (metal ion fraction activity values) are presented in Table 22 [15]; the data of contamination of bone samples of various origin in Table 23 [15].

Year	Calf	Young cattle
1962	4.1	2.9
1963	6.7	2.7
1964	5.2	3.6
1965	6.2	3.7
1966	4.5	2.4
1967	2.8	1.0
1968	2.5	2.8
1969	2.5	1.9
1970	1.2	1.5

Table 22

Activity of metal ion fraction in cattle bones between 1962 and 1970 in pCi/g bone [15]

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	-
Young cattle				
metacarpus	1971	3.2	6.2	-
	1972	4.8	3.6	25.4
	1973	4.7	3.5	-
femur	1971	6.4	5.9	-
	1972	4.3	2.9	22.4
	1973	4.1	3.2	-
costa	1971	-	-	-
	1972	5.0	3.5	22.9
	1973	5.0	4.2	-
Sheep bone	1971	-	-	-
	1972	6.0	4.5	33.6
	1973	4.8	3.4	-
Game bone	1971	-	-	-
	1972	8.3	7.2	43.7
	1973	10.3	7.8	-

Table 23

Activity of bone samples of various origin in the years 1971-1973 [15]

It may be stated that the artificial radioactive contamination could be connected chiefly with the age and species of the animal. The contamination of sheep bones is higher than in cattle bones. Presumably this is caused by the different circumstances of keeping, feeding and grazing of the animals.

The contamination of the bones of the animals of various species living among wild conditions is higher than the values measured in domestic animals. The average value is twice as high than the contamination of sheep bones.

5. INCORPORATION STUDIES

About the ^{90}Sr concentration of human bones there are data available from the year 1966. The measured values according to the age groups are shown in Table 24 [20]. The values are within the range 0.24 to 2.27 pCi ^{90}Sr/g Ca; among these the lower values were measured in the older, 20 to 60 years; the higher in the younger, 0 to 5 years age groups.

^{137}Cs incorporation can be detected in the human body as a result of the air pollution due to nuclear tests. The doses originating from this source, based upon the whole-body counting measurements between 1964 and 1973 are demonstrated on Table 25 [21, 22]. According to the calculations 86.1 pCi ^{137}Cs/kg body weight would cause 1 mrem/year dose burden. The 318 pCi/kg measured in men in 1964 decreased till 1973 to 31.8 pCi/kg ^{137}Cs load, thus by one order of magnitude. In women there was a nearly similar change of this value from 198 to 23.2 pCi/kg.

Age groups, year	*Number of samples*	*Mean*	*Minimum*	*Maximum*
0	*5*	*1.45*	*1.18*	*1.45*
0 - 5	*3*	*2.09*	*1.96*	*2.27*
5 - 20	*6*	*1.01*	*0.7*	*1.21*
20 - 60	*41*	*0.55*	*0.24*	*0.75*

Table 24

^{90}Sr content of human bones (tibia and femur) in pCi/g Ca [20]

Year	*1964*	*1965*	*1966*	*1967*	*1968*	*1969*	*1972*	*1973*
Women	*198.0*	*163.6*	*103.3*	*52.5*	*41.3*	-	*24.1*	*23.2*
Men	*318.6*	*258.3*	*155.0*	*77.5*	*46.5*	*42.2*	*32.7*	*31.8*

Table 25

Average yearly dose burden from ^{137}Cs of the adult population of Budapest in pCi/kg

The data are based on the whole-body counting of 5 persons each [21, 22]

6. SUMMARY

In Hungary the measurements of radioactive pollution are carried out for about 20 years and started with the examination of atmospheric pollution. In the course of the years - similarly to other countries - the sphere of sampling and of the investigations have been extended.

This Bulletin is supported mainly by the data which were already published. The omission or absence of data does not mean implicitly the interruption or discontinuation of the measurements in the given institute, only the lack of publications. Nevertheless, we endeavoured to make available the data from the most important topics up to 1973 in the present publication even on the basis of personal communications or literary data still in press.

The evaluation and comparison of the results of the early period are rendered somewhat difficult by the difference of methods and exactitude of the measurements carried out in the individual institutes. However, the amount of pollution or its changes in time recorded in the single series of data still supplies useful information.

It might be explained as well by the limited possibilities of that period that the majority of the data concerns only the total beta-activity (residual beta) or with more developed techniques the activity of the metal-ion fraction.

Therefore among the 45 tables and figures there are only 10 which contain data concerning the activity of the individual nuclides, e.g. ^{90}Sr, ^{137}Cs, ^{85}Kr or total gamma-activity.

The present publication is intended in the first place to supply summarising information about environmental radioactive pollution in Hungary during the last 20 years. The errors of the data taken from the original publications which originate by all means from the not up-to-date methods of the present time, could be adequately evaluated by the specialists, by whom the difficulties concerning the measurements, particularly during the early period, are well-known.

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

Presently by the improvement of measuring methods and by utilizing the internationally compared samples, a well coordinated system may be developed which is more specialised for the control individual nuclides. Its development in Hungary is necessary all the more, since for the putting into opera-

tion the planned nuclear power plant the knowledge of the zero-level of the environment is an indispensable requirement.

In the further volumes of our Bulletin according to our plans we intend to publish in addition to the summarising informations also the newly measured data of environmental radioactivity concerning Hungary.

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