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THE STUDY OF THE TRACE ELEMENT IN ORGANISMS BY NEUTRON ACTIVATION ANALYSIS. I. MULTI-ELEMENT INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF CANNABIS

by

Masaki SHINOGI and others

Yakugaku zasshi, v.94 no.12, pp.1550-1559, December 1974

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THE STUDY OF THE TRACE ELEMENT IN ORGANISMS BY NEUTRON ACTIVATION ANALYSIS. I. MULTIELEMENT INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF CANNABIS

Bowen cited in his book[3] many papers published up to 1965 on the relationship between organisms and trace elements. On the other hand, from about 1970 onwards, attention has been given to the field called bioinorganic chemistry which deals with boundary territory between inorganic chemistry and biological chemistry.

A large number of trace elements are generally expected to be present in organisms and about 60 trace elements including 16 bioelements (bioelements in plants) have been detected from plants and reported to date[4]. This reminds one of the Noddack's "Guesetz der Allgegenwart der Elemente"[5] which states that all the elements are present in all the rocks and minerals found on earth. In 1972, Morrison et al.[6] wrote that plants need traces of Fe, Cu, Zn, Mn, B, Na, Co, Mo and V while animals need Fe, I, Ca, Zn, Mn, Co, Mo, and Se and, in addition, F, B, Ba and Sr and also that many of such elements may exert harmful effects if present in excess.

For analysis of such a large number of elements, activation analysis is the most suited and the only method available. In recent years, application of this method to biological samples has grown rapidly and numerous reports are known on activation analysis of plants. Takeo et al.[7] irradiated tea leaves with neutron, separated the irradiated leaves into five fractions by ultracentrifugation, determined Cu, Mn, Br, Na and K on each fraction and studied the complexes between Cu or Mn and protein. Trace elements Cu and Zn play an important role in plant growth and Souliotis[8] determined them in 8 plants including olive and maize.

Fourcy et al.[9] reviewed studies of the inorganic components of plants by activation analysis while there is a report on determination of 15 elements such as Mn, K and Cu in plant tissues by non-destructive activation analysis with the use of a high-resolution Ge(Li) detector[10]. Pappas et al.[11] analyzed Au and rare earth elements in opium originating from four countries in 1963 and found that the content of these elements differs characteristically from region to region. On the other hand, Perkons et al.[12] and Rayudu et al.[13] analyzed 30 kinds of opium sampled in 20 countries, determined 13 elements and estimated the place of origin from the relative amounts of these elements.

Recently, Mo-Hsiung Yang et al.[14] determined 13 elements in 8 kinds of tobacco leaves originating from three countries and discussed the possibility of estimating the place of origin from the differences in mercury content.

As described above, qualitative and quantitative analyses of trace elements furnish interesting data to those concerned. We are conducting a series of studies on the content of trace elements in medicinal plants which are not only interesting from the standpoint of food chain (inorganic element cycle) but also important pharmacologically. In this paper, we have examined the optimal conditions for non-destructive neutron activation analysis of hemp or Cannabis which is a hallucinogen known as marijuana and which is posing a serious problem of abuse because of its relatively easy availability and report on some knowledge we thereby gained.

Experimental

In order to carry out simultaneous determination of a variety of trace elements on numerous samples, the non-destructive thermal neutron activation analysis which can determine all the detectable elements with one standard sample was applied in accordance with the monostandard method[15-18].

Processing of data and qualitative and quantitative analyses were handled by an electronic computer (OKITAC Model 5090H) and the results were analyzed by the method developed by Takeuchi et al.[15].

Preparation of Samples and Standard Samples

Two whole Cannabis plants, 130 cm and 140 cm in total length, cultivated in Maizuru and sampled in September 1972 were each divided into five sections: upper leaves which are new leaves on the top, middle leaves which correspond to the upper portion of the remaining stem, lower leaves which correspond to the lower portion of the remaining stem, stem bark and root. The soil adhering to the root was also taken as reference sample.

The leaves and root samples were cleansed ultrasonically in purified water for 30 seconds, jet-washed with distilled water, freeze-dried, ground to less than 80 mesh in an agate mortar and thoroughly mixed. The stem was cleansed and dried likewise and cut into small pieces. The soil was dried in air and ground to less than 80 mesh in an agate mortar. Since each sample was stored in powder, the condition of drying at the time of weighing varied from sample to sample. Therefore, all the samples were freshly dried in a constant temperature electric oven at 45° for 45 hours, each sample was weighed accurately and heat-sealed into a well-cleansed polyethylene bag measuring 4 x 3 cm and the sealed sample was further sealed into another polyethylene bag measuring

6 x 4 cm. The irradiation samples weighed about 350 mg each (the root sample weighed 100 mg) and were prepared in duplicate for 1-minute and 5-minute irradiations.

As standard samples of the monostandard method, Au was used for short half-life nuclides (half-life, 1 day or less) while Co was used for long half-life nuclides (half-life, more than 1 day). These standard samples were prepared readily by cutting a disc, 1 cm in diameter, from a 0.1 mm thick foil of Al-Au alloy (0.1% by weight of Au) or Al-Co alloy (2.0% by weight of Co), products of Belgium Bureau Centrale de Mesures Nucleaires. The Au weighed about 24 μg while the Co weighed about 480 μg.

Irradiation with Neutron

The sample was placed in an irradiation capsule together with the standard sample and irradiated for 1 minute, 5 minutes and 1 hour in No.1 pneumatic tube (thermal neutron flux, 1.9×10^{13} neutrons/cm^2/sec) at the Nuclear Reactor Laboratory of Kyoto University. The 1-minute and 5-minute irradiation samples were grouped by series after measurement and subsequently used as samples for 1-hour irradiation.

Gamma-ray Spectrometry

Upon completion of irradiation, the samples were withdrawn from the capsule, taken out of the outer bag and directly submitted to counting of γ-ray without performing any chemical separation. The samples which had been irradiated for 1 minute or 5 minutes for measurement of short half-life nuclides were counted for 200 seconds after a decay of 3 minutes and further counted for 400 seconds after a decay of 30 minutes. The samples which had been irradiated for 1 hour for measurement of long half-life nuclides were counted for 1K second after a decay of 3 days, for 4K seconds after a decay of 1 week, for 8K seconds after a decay of 2 weeks and further for 20K seconds after a decay of 1 month. The apparatuses used were a 4096 channel pulse height analyzer (manufactured by Nuclear Data) equipped with a 24.7 ml coaxial Ge(Li) detector (manufactured by ORTEC) and, for measurements after a decay of 1 month, a 4096 channel pulse height analyzer (manufactured by Northern Scientific) equipped with a 42.7 ml coaxial Ge(Li) detector (manufactured by ORTEC). A polymethylmethacrylate plate, 1 cm in thickness, was used as a β-ray absorber.

Typical gamma-ray spectra obtained are shown in Figs. 1-4.

Results

The nuclides detected from the relationship between irradiation time and cooling time are shown in Table 1.

For determination of any nuclide which emits two or more γ-rays differing in energy or which is detected twice or more at different times of measurement, the γ-ray energy which yields the smallest errors in measurement was adopted. A total of 41 elements detected, including 35 elements detected in Cannabis (leaves, stem bark and root), and their elemental concentrations are shown in Table 2.

Discussion

Optimal experimental conditions must be found for simultaneous determination of a variety of trace elements on a large number of samples by non-destructive activation analysis and the above-mentioned experimental results were carefully examined to derive such optimal conditions.

Preparation of Samples

a) Cleansing of Samples

No detailed reports are available on cleansing of samples in activation analysis. At any rate, perfect cleansing is extremely difficult to perform as there is always a possibility of some components eluting out in the course of excessive cleansing or of some contaminants causing errors in measurement as a result of insufficient cleansing. Hence, it is necessary to choose optimal conditions for each sample. This time, the ultrasonic cleansing method was investigated on a large number of plant samples for the purpose of removing dusts adhering to both sides of leaves. The leaves of brownish white kidney beans which had been cultivated by hydroponics for two weeks and then allowed to absorb ^{60}Co from the root for two days were used as sample. The sample and 100 ml of purified water were placed in a beaker and subjected to ultrasonic cleansing for 30 seconds or 60-seconds: 5 ml of the water was sampled and measured for radioactivity by a well-type NaI(Tl) scintillation counter.

It was found that no ^{60}Co was eluted at all from the leaves in 30 second cleansing while 3-4% of ^{60}Co was eluted in 60-second cleansing. Therefore, on the assumption that there is virtually no difference in strength of cell surface between Cannabis and kidney beans, ultrasonic cleansing for a duration of 30-seconds or so can be utilized in the present case.

When the surface of Cannabis leaves was coated with talc which had adsorbed ^{60}Co, dried well and cleansed, the talc on the surface was found to have been removed nearly completely by jet-washing with distilled water after 30 second ultrasonic cleansing.

b) Drying

In order to prevent loss by evaporation of specific elements (Hg, Br, As, Se, S), freeze-drying which is regarded most suited for drying of biological samples was applied.

c) Grinding

It is generally considered better to avoid grinding. Here, however, the quantity of sample to be used was small and the sample was ground to less than 80 mesh in an agate mortar and mixed thoroughly to minimize sampling errors. For prevention of external contamination, the sieve used was made of a wooden frame and nylon screen and a pair of tweezers made of Chemifuron was used.

d) Weight of Irradiation Sample

The weight of irradiation sample varies with the irradiation conditions. Under the present conditions, the optimal weight was found to be about 350 mg for 1-minute irradiation in the case of leaves; this weight can be reduced somewhat for 5-minute irradiation as the total induced radioactivity becomes about four times that of 1-minute irradiation. The optimal weights of the stem and root samples were 300 mg and 100 mg respectively for 5-minute irradiation. With the soil sample, the radioactivity of ^{28}Al becomes strong in short-time irradiation when the weight is 350 mg and a distance of one meter had to be allowed for counting of radioactivity after a decay of 3 minutes. Therefore, it is better to take 50 mg or less of the sample in this instance. For long-time irradiation, a weight of 350 mg or so was adequate for measurements after a decay of 1 week or 1 month. It is thus necessary to vary the weight of sample depending upon the length of irradiation time.

Irradiation Time

As for short-time irradiation for measurements of short half-life nuclides, a study was made on 1-minute and 5-minute irradiations.

The gamma-ray spectra presented in Fig. 5 do not show differences in kind of elements detected between 1-minute irradiation and 5-minute irradiation; however, a comparison of photopeak areas of short half-life nuclides in Table 3 indicates that ^{49}Ca, ^{27}Mg or ^{56}Mn yields a larger area after 5-minute

irradiation whereas ^{28}Al or ^{66}Cu yields a larger area after 1-minute irradiation. This is likely due to the Compton peak of Mn which is present in large quantities in the sample becoming larger in 5-minute irradiation and reducing the peak area of the latter. It is further conceivable that short half-life trace nuclides of low energy may sometimes not be detected.

Since the total induced radioactivity after 5-minute irradiation becomes larger than that after 1-minute irradiation, the short-time irradiation should optimally be carried out for 1 minute also from the standpoint of reducing the exposure dose of the experiments at work.

Cooling Time

For measurements of short half-life nuclides by short-time irradiation, it is recommended to make measurement immediately after completion of irradiation and, whenever the total induced radioactivity is large, to make measurement immediately at a greater distance instead of waiting for some decay to occur. However, in the present experiment, a period of about 3 minutes was required between completion of irradiation and start of measurement for transport of the sample and exchange of the outer bag.

It was expeted that, in measurements after a decay of 30 minutes, the decay of ^{28}Al (T = 2.3 min) allows detection of peaks of other nuclides which have otherwise been masked by ^{28}Al and, in addition, produces smaller errors. However, short half-life trace nuclides such as ^{51}Ti (T = 5.8 min), ^{52}V (T = 3.7 min) and ^{66}Cu (T = 5.1 min) were found to have decayed too far to be detected. Therefore, in order to study the cooling time of short half-life nuclides whose half-life is less than 10 minutes, the decay of the induced radioactivity for short half-life nuclides relative to that of ^{28}Al after 1-minute irradiation is shown in Fig. 6 with the average elemental composition of plants[19] as standard.

It is apparent from Fig. 6 that ^{28}Al decayed sufficiently in 30 minutes but other nuclides except ^{49}Ca and ^{27}Mg decayed likewise and this did not permit measurements. In 10 minutes or so, however, ^{28}Al decays to about 1/10 of the level after 3 minutes and loses some of its influence on other nuclides and this is expected to enable measurements of those nuclides which could not be measured after 3 minutes. The above-mentioned results indicate that measurements after 10 minutes instead of 30 minutes are suitable as preliminary measurements after 3 minutes.

In measurements of long half-life nuclides, the cooling times of 3 days and 1 week were studied for those nuclides whose half-life is 2 days or less.

The gamma-ray spectra shown in Fig. 7 indicate that the background by the Compton peaks of ^{24}Na and ^{42}K is large after 3 days due to the presence of large quantities of Na and K in the sample of the S/N ratio of ^{82}Br (T = 35.5 hr) or ^{140}La (T = 40.2 hr) to be measured here becomes small resulting in larger experimental errors. After 1 week, both ^{24}Na (T = 15.0 hr) and ^{42}K (T = 12.5 hr) decayed to such an extent that their effects became insignificant and the experimental errors became smaller. Furthermore, it becomes possible to determine those nuclides which have not been detected after 3 minutes. Thus, measurements after a decay of 1 week were found more effective in this instance.

With respect to nuclides whose half-life is 2 days or more, the gamma-ray spectra shown in Fig. 8 indicate that nuclides such as ^{82}Br, ^{140}La and ^{153}Sm still remain after 2 weeks.

These nuclides emit a large quantity of gamma-ray thereby interfering with other nuclides and causing larger errors in measurement. In 1 month, however, ^{92}Br, ^{140}La and ^{153}Sm decay and their interference disappears. Hence, it is better to make measurements after 1 month in this case.

A method for non-destructive, simultaneous determination of a large number of elements present in plants by activation analysis was established by careful examination of the above-mentioned experimental conditions.

Results of Determination

The contents of elements detected in leaves, root and soil are shown in Fig. 9 in terms of the root/soil ratio and the leaves/root ratios.

Any of the leaves/root ratios is greater than the root/soil ratio for Ca, Mn, K, Ba, Mg, Rb and Zn; that is, these elements migrate from root to various parts of leaves and accumulate there more than they are absorbed from the soil by root. On the other hand, any of the leaves/soil ratios is smaller than the root/soil ratio for Sb, Co, Ce, Eu, Se, Au, Al and Fe, which indicates that these elements are absorbed by the root and accumulate there more than they migrate to various parts of leaves.

Comparison of the elements detected with the afore-mentioned average elemental composition of plants[19] reveals the absence of any significant difference between the two; in particular, a higher content of Hg in Cannabis may be accounted for by agricultural chemicals.

In the present study, the experiments were carried out for the purpose of establishing the experimental conditions whereby the number of samples used was small and no accurate information was obtained on the time of sampling, difference in sex and the like. Hence, no further consideration of the experimental results can be expected. Finally, of the bioelements in plants, the following were not detected this time: H, O, B, C, N, P, S and Mo. H and C cannot be detected by thermal neutron activation analysis. On the other hand, B, N and O present technical difficulties in measurement since the half lives of the nuclides produced are extremely short as follows: $^{11}B(n,\gamma)^{12}B$ (T = 0.02 sec), $^{15}N(n,\gamma)^{16}N$ (T = 7.14 sec) and $^{18}O(n,\gamma)^{19}O$ (T = 29.1 sec).

S is small in both abundance and activation cross section and, although the induced radioactivity is extremely small, S undergoes the following nuclear reactions: $^{36}S(n,\gamma)^{37}S$ (T = 5.0 min) and $^{34}S(n,\gamma)^{35}S$ (T = 87.9 days). Of these, the gamma-ray energy of ^{37}S or 3.102 MeV coincides with that of ^{49}Ca (T = 8.8 min) or 3.084 MeV; moreover, ^{37}S has a short half-life and, even if separated chemically, it is measured with difficulty and hence undetectable. On the contrary, ^{35}S does not emit γ-ray but has a long half-life and it can be determined by chemical separation followed by measurement of β-ray.

Likewise, P can be determined by measuring β-ray from ^{32}P produced by the reaction $^{31}P(n,\gamma)^{32}P$ (T = 14 days), but this requires chemical separation.

On the other hand, Mo undergoes the following reactions and each emits a lot of γ-ray: $^{92}Mo(n,\gamma)^{93m}Mo$ (T = 6.95 hr), $^{98}Mo(n,\gamma)^{99}Mo$ (T = 69.7 hr) and $^{100}Mo(n,\gamma)^{101}Mo$ (T = 14.6 min). Now, it should be possible to determine ^{93m}Mo and ^{101}Mo by 1-minute irradiation followed by counting after a decay of 3 minutes or 30 minutes but such determinations are impossible in actuality on account of a large interference by the Compton peaks of ^{28}Al and ^{56}Mn. Moreover, it should be possible to detect ^{99}Mo by 1-hour irradiation followed by counting after a decay of 1 week but such was not possible this time due to interference by γ-ray from ^{32}Br and ^{140}La. However, the determination would be possible if ^{99}Mo with a long half-life were chemically separated after 1-hour irradiation and its γ-ray of 740 keV measured.

In consequence, S, P and Mo which are important bioelements of plants and which could not be detected this time can be determined by activation analysis with simultaneous use of chemical separation and a method for analysis of these elements will be studied in the future.

Acknowledgement

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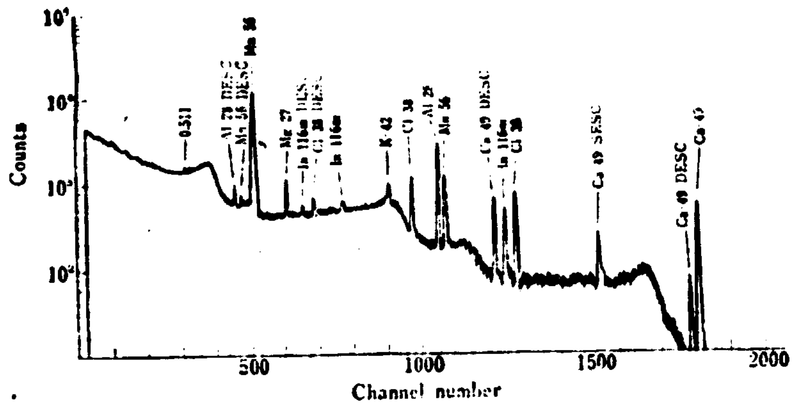

Fig. 1. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 1 Minute and counted for 3 Minutes after a Decay of 3 Minutes

(SESC; single escape peak DESC; double escape peak)

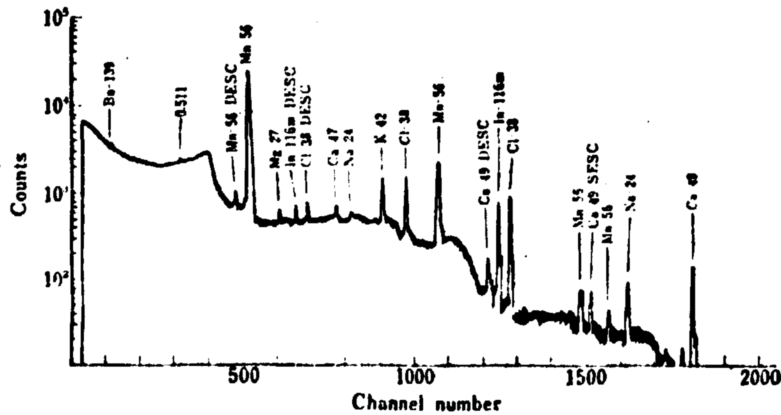

Fig. 2. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 1 Minute and counted for 400 Seconds after a Decay of 30 Minutes

(SESC; single escape peak DESC; double escape peak)

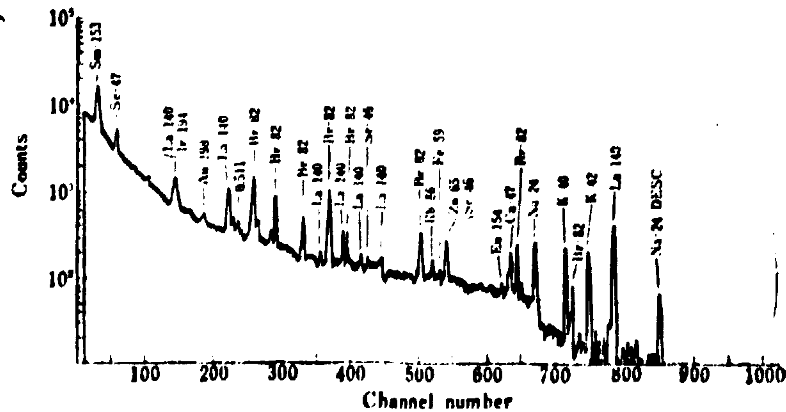

Fig. 3. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 60 Minutes and counted for 4K Seconds after a Decay of 7.7 Days

(DESC; double escape peak)

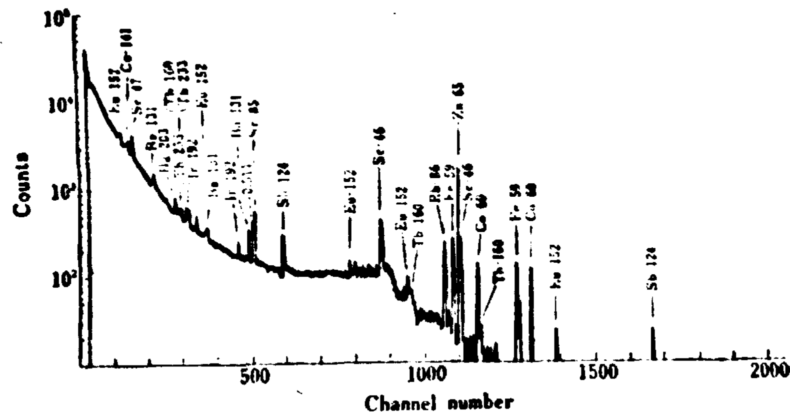

Fig. 4. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irraniated for 60 Minutes and counted for 20K Seconds after a Decay of 32.8 Days

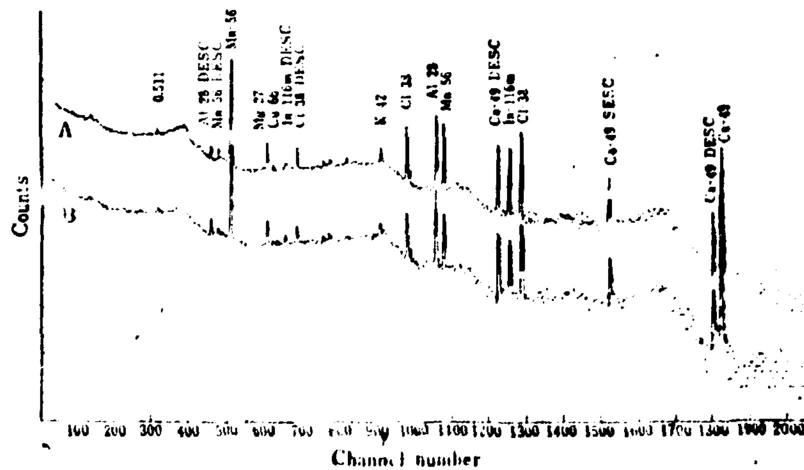

Fig. 5 Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 1 Minute and 5 Minutes, and counted for 3 Minutes after a Decay of 3 Minutes

(SESC, single escape peak DESC, double escape peak)
Irradiation time
A: 5 min
B: 1 min

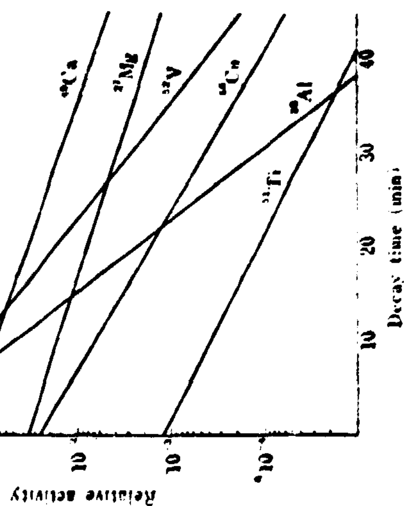

Fig. 6. Induced Radioactivity and Decay Against ^{28}Al in Short Half-lived Nuclides in Plants by 1 Minute Irradiation

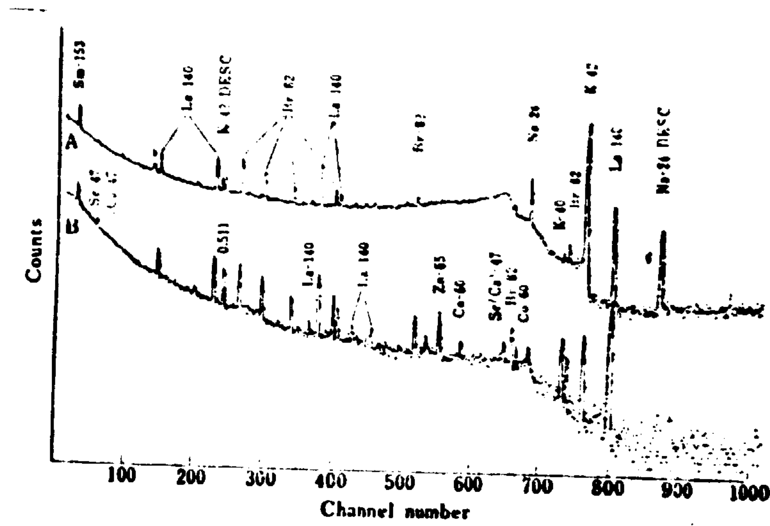

Fig. 7. Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 60 Minites and counted after a Decay of 3 Days and 7 Days

(DESC; double escape peak)
decay time
A: 3 days
B: 7 days

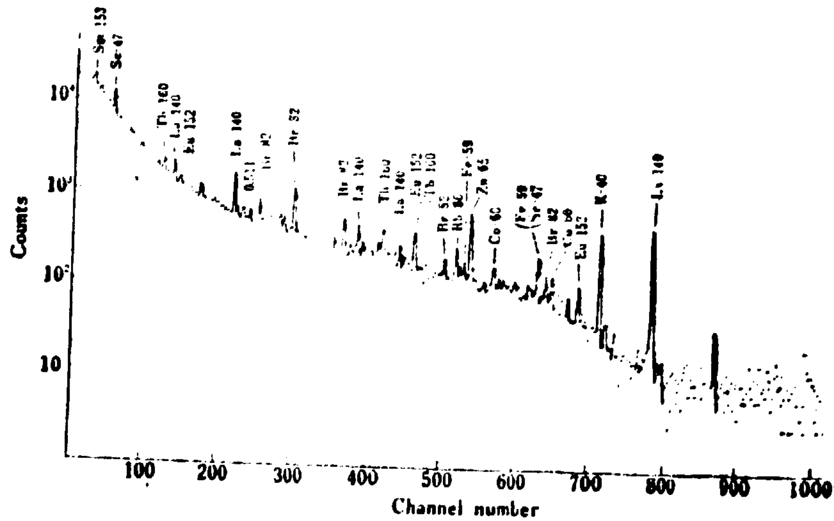

Fig. 8. Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 60 Minutes and counted for 10K Seconds after a Decay of 2 Weeks

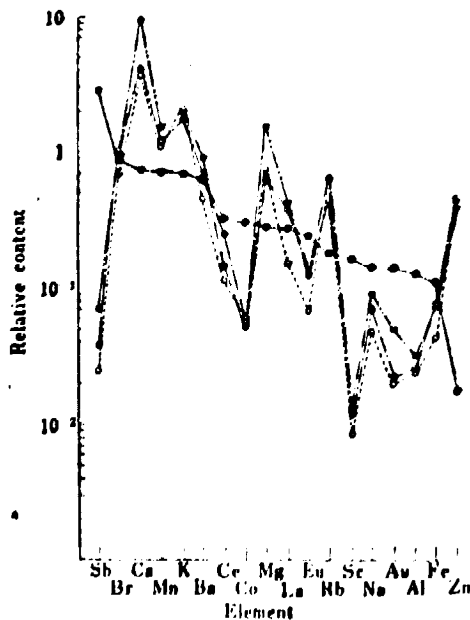

Fig. 9. Relative Content of Each Element in Cannabis (root/soil ratio and various parts of leaves/root ratio)

●—●: root/soil ratio
○---○: upper leaves/root ratio
◐—◐: middle leaves/root ratio
⊖---⊖: lower leaves/root ratio

TABLE I. Nuclear Data for Elements Determined in Cannabis

Measurement	Irradiation time	Decay time	Count interval	Radioisotope measured	Half-life	Best γ-ray energy used (MeV)
1.	1.0 min and 5.0 min	3.0 min	3.0 min	^{28}Al	2.31m	1.779,
				^{139}Ba	82.9m	0.166
				^{49}Ca	8.80m	3.084
				^{38}Cl	37.3m	1.643, 2.168
				^{66}Cu	5.15m	1.039
				^{165}Dy	139m	0.095
				^{116m}In	54.2m	1.293, 2.111
				^{27}Mg	9.45m	1.014
				^{56}Mn	2.58 h	0.847, 1.811
				^{51}Ti	5.80m	0.320
				^{52}V	3.76m	1.434
2.	1.0 min and 5.0 min	30 min	400 sec	^{139}Ba	82.9m	0.166
				^{38}Cl	37.3m	1.643, 2.168
				^{128}I	25.0m	0.443
				^{116m}In	54.2m	1.293, 2.111
				^{42}K	12.5 h	1.525
				^{27}Mg	9.45m	1.014
				^{56}Mn	2.58 h	0.847, 1.811
				^{24}Na	15.0 h	1.369
3.	60 min	1 week	4 K sec	^{198}Au	2.70 d	0.412
				^{82}Br	35.5 h	0.777, 1.317
				^{115m}Cd	43.0 d	0.485
				^{141}Ce	33.0 d	0.145
				^{192}Ir	74.2 d	0.317, 0.468
				^{140}La	40.2 h	0.487, 1.596
				^{24}Na	15.0 h	1.369
				^{46}Sc	83.9 d	0.889, 1.121
				^{153}Sm	47.0 h	0.103
				^{86}Rb	18.7 d	1.079
4.	60 min	1 month	20 K sec	^{110m}Ag	253 d	0.658
				^{131}Ba	12.0 d	0.496
				^{141}Ce	33.0 d	0.145
				^{60}Co	5.26 y	1.173, 1.333
				^{51}Cr	27.8 d	0.320
				^{134}Cs	2.05 y	0.796
				^{152}Eu	12.7 y	0.344, 1.408
				^{59}Fe	45.0 d	1.099, 1.292
				^{153}Gd	242 d	0.097
				^{181}Hf	42.5 d	0.482
				^{203}Hg	46.9 d	0.279
				^{192}Ir	74.2 d	0.317, 0.468
				^{177}Lu	6.74 d	0.208
				^{86}Rb	18.7 d	1.079
				^{124}Sb	60.4 d	0.603, 1.691
				^{46}Sc	83.9 d	0.889, 1.121
				^{182}Ta	115 d	1.189
				^{160}Tb	72.1 d	0.879
				^{233}Th	27.0 d	0.312
				^{169}Yb	32.0 d	0.198
				^{65}Zn	245 d	1.115

Table II. Elemental Concentration in Cannabis Bred in Maizuru (ppm)

	Leaves			Stem bark	Root	Soil
	Upper	Middle	Lower			
Ag	—	—	0.12	—	—	—
Al	130	140	170	68	5300	41000
Au	0.0020	0.0023	0.0051	0.0014	0.10	0.69
Ba	46	70	92	90	100	160
Br	2.0	2.9	2.7	1.1	2.8	3.2
Ca	21000	24000	54000	9700	5700	7600
Cd	—	—	510	2.2	—	—
Ce	0.87	1.9	1.1	0.13	7.5	23
Cl	1500	1800	1800	2800	230	—
Co	0.11	0.11	0.099	0.32	1.9	6.1
Cr	—	—	—	—	—	80
Cs	a)	0.011	0.013	—	0.26	3.0
Cu	6.0	13	300	—	—	—
Dy	—	—	—	—	0.39	—
Eu	0.0082	0.016	0.015	—	0.12	0.49
Fe	130	220	210	53	2900	26000
Gd	—	—	—	—	19	140
Hf	—	—	—	—	—	3.4
Hg	0.093	0.14	0.30	0.055	—	—
I	—	—	—	—	5.3	—
In	0.11	0.14	2.4	0.20	0.24	a)
Ir	0.000032	0.00012	0.000066	0.00027	—	—
K	20000	17000	19000	22000	9600	14000
La	0.61	1.5	1.7	0.12	3.9	14
Lu	—	—	—	—	—	0.47
Mg	5000	4100	10000	2200	6500	23000
Mn	110	120	150	54	100	140
Na	46	68	88	210	940	6500
Pr	—	—	—	430	—	—
Rb	4.4	5.6	3.7	5.5	8.3	45
Ru	—	—	a)	1.3	—	—
Sb	0.035	0.055	0.10	0.61	1.4	0.50
Sc	0.012	0.017	0.18	0.016	1.4	8.3
Sm	0.54	0.16	0.18	—	—	6.1
Ta	—	—	—	—	—	0.28
Tb	0.0067	0.013	a)	a)	a)	0.91
Th	—	—	—	—	—	5.0
Ti	—	—	—	—	580	4500
V	—	—	—	—	8.7	61
Yb	—	—	—	—	—	0.41
Zn	38	40	33	41	84	4700

a) only qualitative

Table III. Comparison of Photopeak Area of Short Half-lived Nuclides in Middle Leaves of Cannabis by Irradiated for 1 Minute and 5 Minutes
Unit in counts/minute/gram

Nuclide	γ-Ray energy (MeV)	Irradiation time	
		1 minute	5 minutes
^{28}Al	1.779	8710	7770
^{66}Cu	1.039	190	172
^{49}Ca	3.084	2370	3220
^{27}Mg	1.014	1460	1740
^{56}Mn	0.847	3510	5540

YAKUGAKU ZASSHI
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中性子放射化分析による生体中微量元素の研究（第 1 報）
大麻の機器的多元素放射化分析[1]

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The Study of the Trace Element in Organisms by Neutron Activation Analysis. I.
Multielement Instrumental Neutron Activation Analysis of Cannabis

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Examinations were made on optimal experimental conditions for instrumental determination of various elements in cannabis by neutron activation analysis, without any radiochemical separation, and the following conditions were found to be useful. Irradiation samples to be used are about 300 mg of the leaves or stem bark, and about 100 mg of the root. For soil sample, about 50 mg is used for the determination of short half-life nuclides and about 300 mg for long half-life nuclides. For short half-life nuclides, the samples are irradiated for 1 min, activity is measured for 200 sec after a decay of 3 min, and for 400 sec after a decay of 10 min. For long half-life nuclides, the samples are irradiated for 60 min and the activities are measured for 4 K sec after 1 week and for 10 K sec after 1 month. Use of supersonic waves is also convenient for cleansing of the samples. Thirty five kinds of interesting elements were determined by this method from cannabis cultivated in Maizuru area.

生物と微量元素との関係について，Bowen はその著書[3] で 1965 年までの多くの論文を紹介している．一方，1970 年頃から，無機化学と生物化学の境界領域として生物無機化学 (bioinorganic chemistry) とよばれる分野が注目されている．

一般に生体中には数多くの微量元素が存在すると予想され，現在までに一般植物体中から検出され，報告されている元素は，16 の生元素（植物体における生元素）を含む約 60 種あまりであり,[4] このことは，地球上に存在するすべての岩石・鉱物中には，すべての元素が存在するという Noddack の "元素普存説"[5] を想起させる．また，1972 年に Morrison ら[6] は，植物生体に微量の Fe, Cu, Zn, Mn, B, Na, Co, Mo および V が必要であり，動物生体には Fe, I, Ca, Zn, Mn, Co, Mo および Se のほかに F, B, Ba および Sr も必要であると述べているが，しかし多くの元素は必要以上に存在すると有害な影響を与えるとも報じている．

このような多数の元素を分析するには，放射化分析が最も適した方法であり，唯一の手段である．近年，本法の生体試料への応用は急激に増加し，一般植物の放射化分析についても多数報告されている．竹尾ら[7] は，茶葉を中性子照射後，超遠心分離により 5 分画にわけそれぞれの分画について Cu, Mn, Br, Na, K を定量し，Cu および Mn のタンパクとの complex について検討しており，Souliotis[8] は，植物生長に重要な役割をする微量元素 Cu, Zn を，オリーブ，とうもろこしなど 8 種類の植物から定量している．

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1) 日本薬学会近畿支部第 23 回総会で発表，京都，1973 年 11 月．
2) Location: *a) Motoyama-kitamachi, Higashinada-ku, Kobe*; *b) Kumatori-cho, Sennan-gun, Osaka.*

3) H.J.M. Bowen, "Trace Elements in Biochemistry," Academic Press, London and New York, 1966.
4) 服部明彦編，"海洋科学基礎講座 11 巻 海洋生化学，" 東海大学出版会，東京，1973.
5) W.I. Noddack, *Angew. Chem.*, 47, 637 (1934); *idem, ibid.*, 49, 835 (1936).
6) G.H. Morrison, N.M. Potter, Abstracts of Papers, IUPAC, International Congress on Analytical Chemistry, Kyoto, April, 1972, p. 441.
7) T. Takeo, M. Shibuya, *Radioisotopes*, 20, 25 (1971).
8) A.G. Souliotis, *Analyst*, 94, 359 (1969).

Fourcy ら[9] は，放射化分析による植物の無機成分の研究についての応用をまとめており，また高分解能の Ge(Li) 検出器を使用した非破壊放射化分析による Mn, K, Cu など 15 元素を種々の植物体から定量した報告[10] もある．一方，Pappas ら[11] は 1963 年に 4 ヶ国のアヘンについて，Au と希土類元素を分析し，これらの含有量の相対的な相違から地域的な特徴のあることを報告し，Perkons[12] および Rayudu ら[13] は，20 ヶ国から採取した 30 種のアヘンを分析し，13 元素を定量して，それぞれの元素の有無により産地を推定している．最近では Mo-Hsiung Yang ら[14] が 3 ヶ国 8 品種のタバコ葉から 13 元素を定量し，その水銀含有量の相違から産地推定の可能性を論じている．

このように微量元素の定性および定量結果は種々の立場に興味あるデータを提供している．著者らは，食物連鎖（無機元素サイクル）的にも興味ある植物で，しかも薬学的にも重要である薬用植物中の微量元素含有量を明らかにし，種々検討しているが，本報ではマリファナとして知られている幻覚剤で，その入手が比較的容易であるため，乱用が問題となっている大麻の非破壊中性子放射化分析を行なうために最も都合の良い条件を検討し，若干の知見を得たので報告する．

実　験

多数の試料について多種類の微量元素を同時定量するために，1 つの標準試料で検出されるすべての元素を定量することができる，モノスタンダード法[15-18] による非破壊熱中性子放射化分析法を応用した．

なお，データ処理および定性，定量は電子計算機 (OKITAC 5090H 型) を使用し，武内らの方法[15] により解析した．

試料および標準試料の調製　試料は，1972 年 9 月に採取した[illegible]産大麻全長 130 cm と 140 cm の 2 本を，頂上部の新芽部分を上葉部とし，残りの茎の上部を中葉部，下部を下葉部とし，さらに茎の皮および根の 5 部位

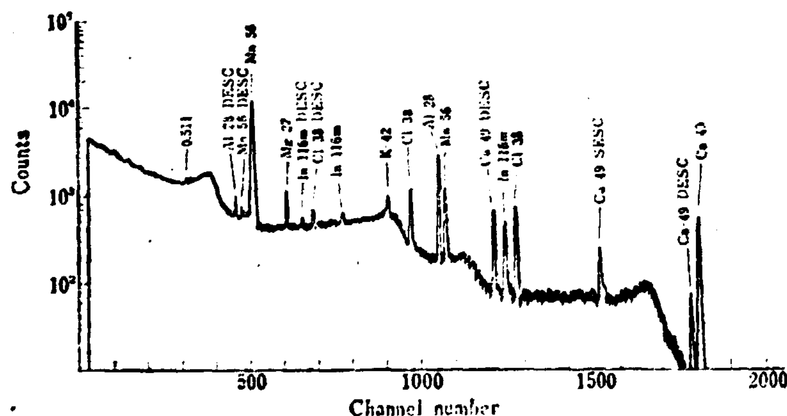

Fig. 1. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 1 Minute and counted for 3 Minutes after a Decay of 3 Minutes

(SESC; single escape peak DESC; double escape peak)

9) A. Fourcy, M. Neuburger, *Bull. Soc. Chim. France*, 11, 4681 (1968).
10) W.A. Haller, L.A. Rancitelli, T.A. Cooper, *J. Agr. Food Chem.*, 16, 1036 (1968).
11) A.C. Pappas, J. Alsted, G. Lunde, *Radiochemica Acta*, 1, 109 (1963).
12) A.K. Perkons, R.E. Jervis, Proc. 1st National Symposium on Law Enforcement Science and Technology, Chicago, March, 1967, p. 257.
13) G.V.S. Rayudu, B. Tiefenbach, R. E. Jervis, Trans. 14th Annual Meeting of the ANS/CNA, Toronto, June, 1968, p. 81.
14) M.-H. Yang, S.-F. Lai, S.-J. Yeh, *Radioisotopes*, 22, 118 (1973).
15) T. Takeuchi, T. Hayashi, *Annu. Rep. Res. Reactor Ins. Kyoto Univ.*, 3, 9 (1970).
16) T. Takeuchi, T. Hayashi, Y. Kusaka, *Annu. Rep. Res. Reactor Ins. Kyoto Univ.*, 4, 63 (1971).
17) T. Takeuchi, T. Hayashi, *Annu. Rep. Res. Reactor Ins. Kyoto Univ.*, 5, 49 (1972).
18) T. Takeuchi, M. Shinogi, *Annu. Rep. Res. Reactor Ins. Kyoto Univ.*, 6, 68 (1973).

に区分した．なお，根に付着した土も参考のために試料とした．

葉および根試料は，精製水による30秒間の超音波洗浄後，蒸留水で噴射洗浄を行ない，凍結乾燥後，メノウ製乳鉢で80メッシュ以下に粉砕し，よく混合した．茎は同様に洗浄，乾燥後，細かく刻んだ．土は風乾後，メノウ製乳鉢で80メッシュ以下に粉砕した．各試料を粉末にして保管しておいたので秤量する時点において乾燥状態が全試料一定ではないので，あらためて全試料を電気定温乾燥器により45°で45時間乾燥した．その後精秤し，よく洗浄した4×3cmのポリエチレン袋に溶封し，さらに6×4cmのポリエチレン袋で2重に封入した．照射試料は約350mg（根は100mg）とし各2個ずつ作製して1分および5分照射の試料とした．

モノスタンダード法の標準試料として，短寿命核種（半減期が1日以内）にはAu，長寿命核種（半減期が1日以上）にはCoを用いた．それぞれ調製の簡単な，Al-0.1% (w/w) Au合金，およびAl-2.0% (w/w) Co合金フォイル（厚さ0.1 mm, Belgium Burean Central de Mesures Nucleaires社製）を直径1 cmの円型に切りぬき使用した．Auの重量は約24 μg，Coの重量は約480 μgであった．

中性子照射　試料をおのおの標準試料とともに照射用カプセルにつめ，京都大学原子炉実験所の圧気輸送管No. 1（熱中性子束 1.9×10^{13} neutron/cm²/sec）において，1分，5分，および1時間照射を行なった．なお，1分および5分照射した試料を測定後，各シリーズにまとめて1時間照射の試料とした．

Fig. 2. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 1 Minute and counted for 400 Seconds after a Decay of 30 Minutes

(SESC; single escape peak DESC; double escape peak)

Fig. 3. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irradiated for 60 Minutes and counted for 4K Seconds after a Decay of 7.7 Days

(DESC; double escape peak)

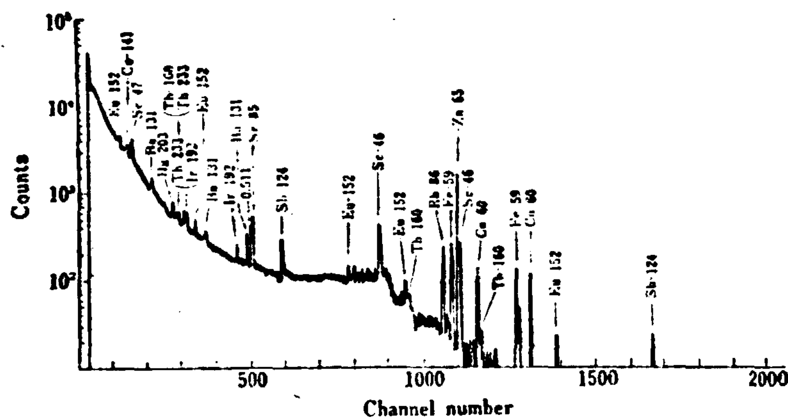

Fig. 4. Typical Gamma-ray Spectra of Upper Leaves of Cannabis irraniated for 60 Minutes and counted for 20K Seconds after a Decay of 32.8 Days

γ線スペクトロメトリー　照射終了後，試料をカプセルからとり出し外袋を取り除いて，どのような化学分離操作も行なわずに直接γ線の測定を行なった．短寿命核種測定のために1分および5分照射した試料は照射終了後，3分後に200秒，さらに30分後に400秒測定した．長寿命核種測定のために1時間照射した試料は，3日後に1K秒，1週間後に4K秒，2週間後に8K秒，さらに1カ月後には20K秒測定した．使用した測定器は，24.7 mlの同軸型Ge(Li)検出器(ORTEC社製)を装備した4096チャンネル波高分析器(Nuclear Data社製)，および1カ月後の測定には42.7 mlの同軸型Ge(Li)検出器(ORTEC社製)を装備した4096チャンネル波高分析器(Northern Scientific社製)を使用し，β線吸収板として，厚さ1 cmのアクリル板を用いた．

得られたγ線スペクトルの代表的な例を，Fig. 1—4に示す．

結　　果

照射時間と冷却時間の関係から，検出された核種の結果をTable Iに示す．

同一元素で，2本以上のエネルギーの異なるγ線を放出するもの，および異なる測定時に2回以上検出された核種の定量には，それぞれ定量誤差の最も小さいγ線エネルギーを採用した．以上の結果，大麻（葉，茎，根）より検出された35元素を含む全検出元素41元素とその定量値をTable IIに示す．

Table I. Nuclear Data for Elements Determined in Cannabis

Measurement	Irradiation time	Decay time	Count interval	Radioisotope measured	Half-life	Best γ-ray energy used (MeV)
1.	1.0 min and 5.0 min	3.0 min	3.0 min	^{28}Al	2.31m	1.779,
				^{139}Ba	82.9m	0.166
				^{49}Ca	8.80m	3.084
				^{38}Cl	37.3m	1.643, 2.168
				^{66}Cu	5.10m	1.039
				^{165}Dy	139m	0.095
				^{116m}In	54.2m	1.293, 2.111
				^{27}Mg	9.45m	1.014
				^{56}Mn	2.58h	0.847, 1.811
				^{51}Ti	5.80m	0.320
				^{52}V	3.76m	1.434

2.	1.0 min and 5.0 min	30 min	400 sec	^{139}Ba	82.9 m	0.166
				^{38}Cl	37.3 m	1.643, 2.168
				^{128}I	25.0 m	0.443
				^{116m}In	54.2 m	1.293, 2.111
				^{42}K	12.5 h	1.525
				^{27}Mg	9.45 m	1.014
				^{56}Mn	2.58 h	0.847, 1.811
				^{24}Na	15.0 h	1.369
3.	60 min	1 week	4 K sec	^{198}Au	2.70 d	0.412
				^{82}Br	35.5 h	0.777, 1.317
				^{115m}Cd	43.0 d	0.485
				^{141}Ce	33.0 d	0.145
				^{192}Ir	74.2 d	0.317, 0.468
				^{140}La	40.2 h	0.487, 1.596
				^{24}Na	15.0 h	1.369
				^{46}Sc	83.9 d	0.889, 1.121
				^{153}Sm	47.0 h	0.103
				^{86}Rb	18.7 d	1.079
4.	60 min	1 month	20 K sec	^{110m}Ag	253 d	0.658
				^{131}Ba	12.0 d	0.496
				^{141}Ce	33.0 d	0.145
				^{60}Co	5.26 y	1.173, 1.333
				^{51}Cr	27.8 d	0.320
				^{134}Cs	2.05 y	0.796
				^{152}Eu	12.7 y	0.344, 1.408
				^{59}Fe	45.0 d	1.099, 1.292
				^{153}Gd	242 d	0.097
				^{181}Hf	42.5 d	0.482
				^{203}Hg	46.9 d	0.279
				^{192}Ir	74.2 d	0.317, 0.468
				^{177}Lu	6.74 d	0.208
				^{86}Rb	18.7 d	1.079
				^{124}Sb	60.4 d	0.603, 1.691
				^{46}Sc	83.9 d	0.889, 1.121
				^{182}Ta	115 d	1.189
				^{160}Tb	72.1 d	0.879
				^{233}Th	27.0 d	0.312
				^{169}Yb	32.0 d	0.198
				^{95}Zr	245 d	1.115

Table II. Elemental Concentration in Cannabis Bred in Maizuru (ppm)

	Leaves			Stem bark	Root	Soil
	Upper	Middle	Lower			
Ag	—	—	0.12	—	—	—
Al	130	140	170	68	5300	41000
Au	0.0020	0.0023	0.0051	0.0014	0.10	0.69
Ba	46	70	92	90	100	160
Br	2.0	2.9	2.7	1.1	2.8	3.2
Ca	21000	24000	54000	9700	5700	7600
Cd	—	—	540	2.2	—	—
Ce	0.87	1.9	1.1	0.13	7.5	23
Cl	1500	1800	1800	2800	230	—
Co	0.11	0.11	0.099	0.32	1.9	6.1

Cr	—	—	—	—	—	80
Cs	a)	0.011	0.013	—	0.26	3.0
Cu	6.0	13	300	—	—	—
Dy	—	—	—	—	0.39	—
Eu	0.0082	0.016	0.015	—	0.12	0.49
Fe	130	220	210	53	2900	26000
Gd	—	—	—	—	19	140
Hf	—	—	—	—	—	3.4
Hg	0.093	0.14	0.30	0.055	—	—
I	—	—	—	—	5.3	—
In	0.11	0.14	2.4	0.20	0.24	a)
Ir	0.000032	0.00012	0.000066	0.00027	—	—
K	20000	17000	19000	22000	9600	14000
La	0.61	1.5	1.7	0.12	3.9	14
Lu	—	—	—	—	—	0.47
Mg	5000	4100	10000	2200	6500	23000
Mn	110	120	150	54	100	140
Na	46	68	88	210	940	6500
Pr	—	—	—	430	—	—
Rb	4.4	5.6	3.7	5.5	8.3	45
Ru	—	—	a)	1.3	—	—
Sb	0.035	0.055	0.10	0.61	1.4	0.50
Sc	0.012	0.017	0.18	0.016	1.4	8.3
Sm	0.54	0.16	0.18	—	—	6.1
Ta	—	—	—	—	—	0.28
Tb	0.0067	0.013	a)	a)	a)	0.91
Th	—	—	—	—	—	5.0
Ti	—	—	—	—	580	4500
V	—	—	—	—	8.7	61
Yb	—	—	—	—	—	0.41
Zn	38	40	33	41	84	4700

a) only qualitative

考　　察

多数の試料について多種類の微量元素を非破壊放射化分析により同時定量するためには適当な実験条件が必要であり，以上の実験結果から，最も良い実験条件を検討し，また，実験結果についても考察した．

試料調製の検討

a. 試料洗浄について　放射化分析における試料洗浄に関し，詳細な報告はない．しかし，洗浄による成分の溶出や，洗浄不足による汚染の誤差が生じる懸念があり，完全な洗浄は非常に困難である．したがって各試料について最適と思われる一定条件を定める必要があり，今回は多数の植物試料について，その葉の表裏に付着したチリを落とす目的で超音波洗浄法を検討した．試料は水耕法により2週間栽培し，さらに2日間 ^{60}Co を根から吸収させた蔓白インゲン豆の葉を使用し，ビーカーに精製水 100 ml と試料を入れ，30秒間および60秒間の超音波洗浄を行ない，この洗浄液を 5 ml 採取して井戸型 NaI (Tl) シンチレーション検出器でその放射能を測定した．

以上の結果から，30秒間の洗浄では葉中の ^{60}Co の溶出はまったくみられず，60秒間では 3—4% の溶出がみられた．したがってこの場合，大麻とインゲン豆との間に細胞表面の強度差がほとんどないものとすれば，30秒程度の超音波洗浄が利用できる．

また，大麻の葉の表面に ^{60}Co を吸着させたタルクをぬり，よく乾燥させて洗浄を行なった結果では，30秒間の超音波洗浄後，蒸留水により噴射洗浄することで表面のタルクはほとんど除かれることが判明した．

b. 乾燥について　特定元素 (Hg, Br, As, Se, S) の揮発損失を防ぐため生体試料の乾燥には最も良いとされている凍結乾燥を用いた．

c. 粉砕について　一般に粉砕はさけた方が良いといわれるが使用する試料の量が少ないので，サンプリング誤差を小さくするため，よく混合する目的でメノウ製乳鉢を用い，80 メッシュ以下に粉砕した。なお，外部汚染を防ぐため篩は木ワク付ナイロン網製，ピンセットはケミフロン製のものを用いた。

d. 照射試料の重量について　照射試料の重量については各照射条件により異なるが，今回の条件のもとで最も都合の良い重量を検討した．葉試料は 1 分照射の場合，約 350 mg で適当であったが，5 分照射では全生成放射能が 1 分照射より約 4 倍大きくなるので，照射重量は多少減じても良く，茎，根試料では，各々 300 mg, 100 mg が適当であった．土試料は 350 mg では短時間照射の場合 ^{28}Al の生成放射能が強くなるので，3 分後の測定において測定距離を 1 m 離さなければならなかった．したがってこの場合 50 mg 以下にした方が良い．なお，長時間照射の場合，1 週間後および 1 ケ月後の測定では 350 mg 程度の量が適当であったため，照射時間の長短によって試料重量を変える必要がある．

照射時間の検討

短寿命核種測定のための短時間照射について 1 分および 5 分で検討を行なった．

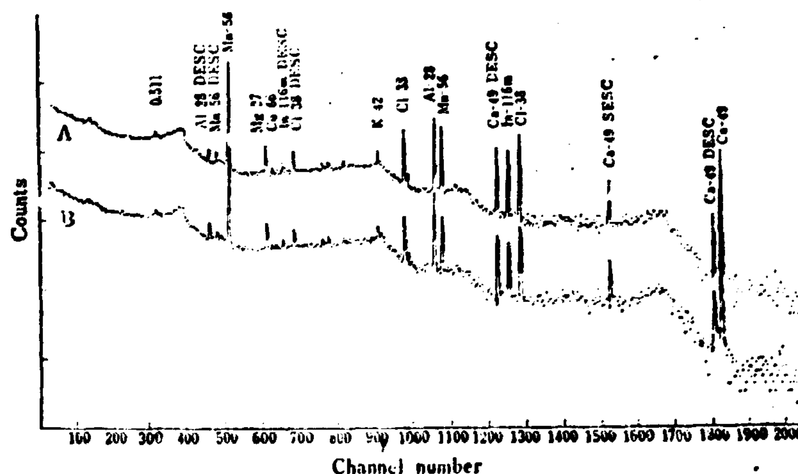

Fig. 5. Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 1 Minute and 5 Minutes, and counted for 3 Minutes after a Decay of 3 Minutes

(SESC; single escape peak DESC; double escape peak)
Irradiation time
A: 5 min
B: 1 min

Table III. Comparison of Photopeak Area of Short Half-lived Nuclides in Middle Leaves of Cannabis by Irradiated for 1 Minute and 5 Minutes
Unit in counts/minute/gram

Nuclide	γ-Ray energy (MeV)	Irradiation time	
		1 minute	5 minutes
^{28}Al	1.779	8710	7770
^{66}Cu	1.039	190	172
^{49}Ca	3.084	2370	2220
^{27}Mg	1.014	1460	1740
^{56}Mn	0.847	3510	5540

Fig. 5 のスペクトルから，検出された元素については，1 分および 5 分照射で相違は認められないが，短寿命核種のピーク面積の比較では，Table III に示すように ^{49}Ca, ^{27}Mg, ^{56}Mn は 5 分照射，^{28}Al, ^{66}Cu は 1 分照射の方が面積は大きくなる．

これは，試料中に多量存在する Mn のコンプトンピークが 5 分照射では大きくなり，後者のピーク面積を減少させるためと考えられ，また，エネルギーの低い短寿命の微量核種も検出されない場合が考えられる．

また，全生成放射能は 5 分照射では 1 分照射より大きくなるので，実験時における実験者の被曝線量を減じるためにも短時間照射は，1 分が適当である．

冷却時間の検討

短時間照射による短寿命核種の測定は，照射終了後ただちに測定を行なうのが良く，全生成放射能が強いときは減衰を待つより，測定距離を離して行なう方が良い．しかし今回は照射終了後，試料の輸送および外袋の交換などのため，測定までに約 3 分を要した．

30 分後の測定は ^{28}Al (T=2.3 分) が減衰し，このためにかくれていた他の核種のピークが検出でき，さらに定量誤差も小さくなるものと予想したが，半減期の短かい ^{51}Ti (T=5.8 分)，^{52}V (T=3.7 分)，^{66}Cu (T=5.1 分) のような微量核種はすでに減衰しており検出できなかった．したがって半減期が 10 分以内の短寿命核種の冷却時間を検討するために，植物体の平均元素組成[19] を基準として，1 分照射時における短寿命核種の ^{28}Al に対する生成放射能およびその減衰の関係を Fig. 6 に示した．

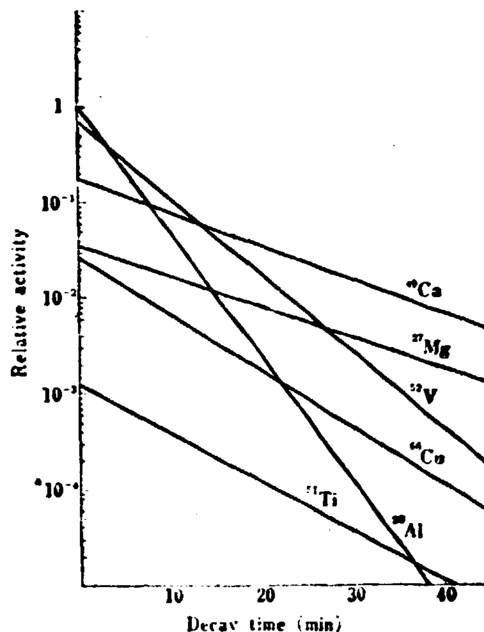

Fig. 6. Induced Radioactivity and Decay Against ^{28}Al in Short Half-lived Nuclides in Plants by 1 Minute Irradiation

この図より，^{28}Al は 30 分後には十分減衰しているが ^{49}Ca, ^{27}Mg 以外の核種も減衰しており測定できない，しかし 10 分程度では ^{28}Al は 3 分後の約 1/10 に減衰し，他の核種への影響も少なくなっているので，3 分後に測定できなかった他の核種の測定が期待できる．以上の結果，30 分後よりも 10 分後に測定する方が 3 分後の予備的測定として適当である．

長寿命核種の測定において，半減期が 2 日以内の核種の冷却時間について，3 日と 1 週間の検討を行なった．

Fig. 7 の γ 線スペクトルから，3 日後では試料中の Na や K の量が多いため，^{24}Na および ^{42}K のコンプトンピークによるバックグラウンドが高く，ここで測定すべき ^{82}Br (T=35.5 時間)，^{140}La (T=40.2 時間) の S/N 比が小さくなり測定誤差が大きくなる．1 週間後では ^{24}Na (T=15.0 時間) や ^{42}K (T=12.5 時間) は共に減衰しており，これらの核種による影響はなく，測定誤差も小さくなる．さらに 3 日後で検出できなかった核種も定量可能となる．したがってこの場合は，1 週間の測定の方が有効であった．

半減期が 2 日以上の核種については，Fig. 8 に示す γ 線スペクトルから，2 週間後でも，まだ ^{82}Br, ^{140}La, ^{153}Sm などの核種が残っている．

これらの核種は多くの γ 線を放出するので他の核種への妨害となり，その定量誤差を大きくする．1 ケ月後では，完全に ^{82}Br, ^{140}La, ^{153}Sm は減衰し，これらによる妨害もなくなる．したがってこの場合は 1 ケ月後に測定を行なう方が良い．

以上の実験条件の考察より，植物体に含まれる多数の諸元素を放射化分析により，非破壊的に同時定量できる方法が確立できた．

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19) Y. Miyake, "Element of Geochemistry," Maruzen, Tokyo, 1965.

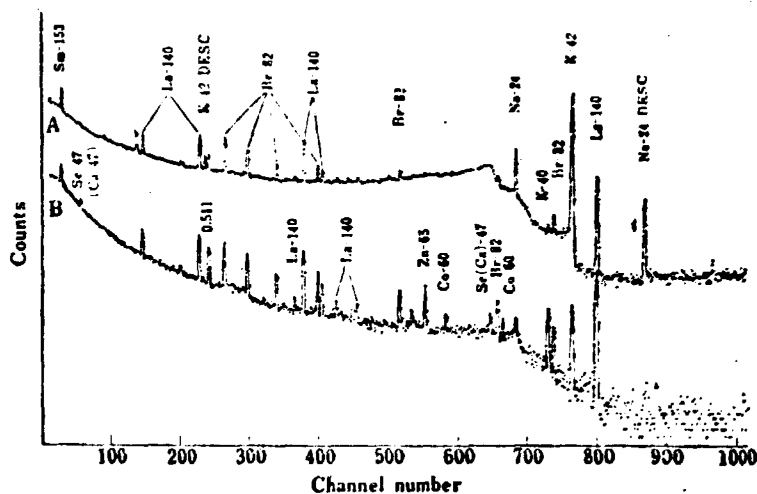

Fig. 7. Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 60 Minites and counted after a Decay of 3 Days and 7 Days

(DESC; double escape peak)
decay time
A: 3 days
B: 7 days

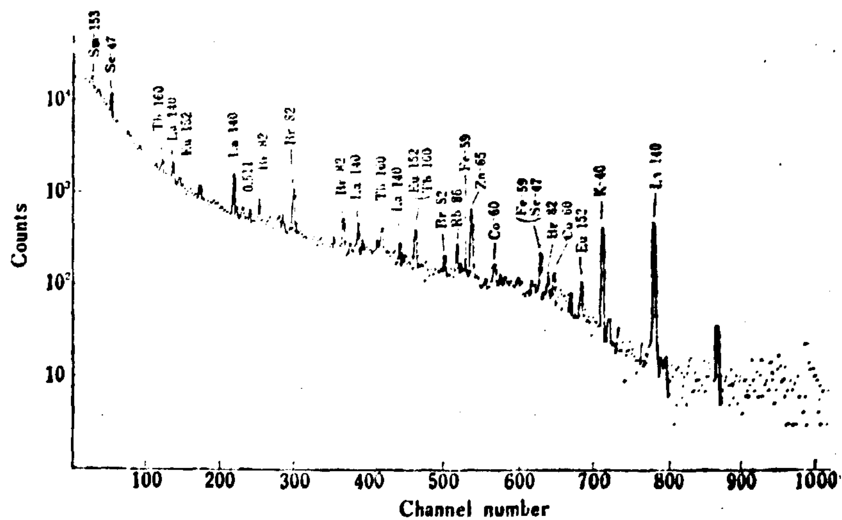

Fig. 8. Gamma-ray Spectra of Middle Leaves of Cannabis irradiated for 60 Minutes and counted for 10K Seconds after a Decay of 2 Weeks

定量結果について

葉の各部位，根および土のすべてにおいて，検出された元素について，その含有量の土に対する根の割合と，根に対する葉の各部の割合を Fig. 9 に示した．

葉の各部/根>根/土の元素として Ca, Mn, K, Ba, Mg, Rb, Zn があり，これらの元素は，根が土から吸収する割合より，根から葉の各部へ移動し，蓄積される割合が大きいことを示している．また，葉の各部/根<根/土

の元素として，Sb, Co, Ce, Eu, Sc, Au, Al, Fe があり，これらの元素は，根にはよく吸収され蓄積されるが葉の各部への移動は小さいことを示している．

なお，検出された各元素について，前述[10]の植物体の平均元素組成との比較では，ほとんど大差はないが，特に Hg については大麻の方が含有量が大きく，これは農薬によるものと考えられる．

本報は実験条件の確立を目的として実験を行なったこと，および使用した試料の数が少ないこと，サンプリング時期，雌雄の別などについての正確な情報が得られなかったので，これ以上の分析結果への考察は期待できない．最後に，植物体における生元素中で，今回検出できなかった元素に H, O, B, C, N, P, S, Mo がある．これらの元素中，H, C は熱中性子放射化分析法では検出できない．また B, N, O については各々 ^{11}B(n, γ) ^{12}B (T=0.02 秒)，^{15}N (n, γ) ^{16}N (T=7.14 秒)，^{18}O (n, γ) ^{19}O (T=29.1 秒) と生成核種の半減期が非常に短いため技術的にも測定は困難である．

S は存在度および放射化断面積がともに小さく，生成放射能は非常に少ないが核反応として次の反応がおこる．^{36}S(n, γ) ^{37}S (T=5.0 分)，^{34}S(n, γ) ^{35}S (T=87.9 日)．このうち ^{37}S の γ 線エネルギー 3.102 MeV は ^{49}Ca (T=8.8 分) の 3.084 MeV と重なり，しかも半減期が短かく化学分離を行なっても測定は困難なため検出できない．しかし ^{35}S は γ 線を放出しないが半減期が長いので化学分離を行ない，その β 線を測定すれば定量が可能となる．

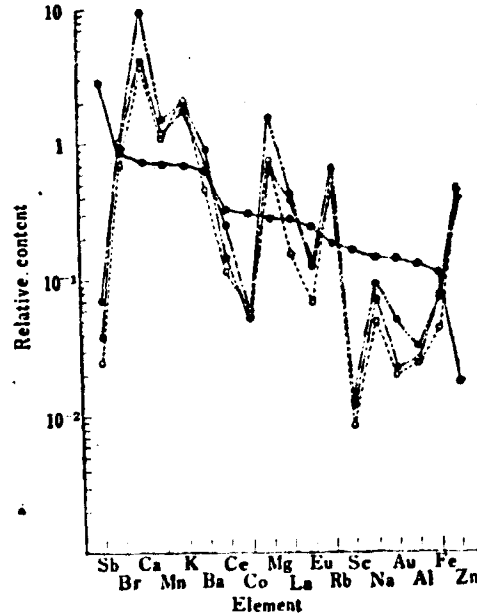

Fig. 9. Relative Content of Each Element in Cannabis (root/soil ratio and various parts of leaves/root ratio)

P についても ^{31}P (n, γ) ^{32}P (T=14 日) によって生成した ^{32}P の β 線を測定すれば定量できるが化学分離の操作を必要とする．

一方，Mo は ^{92}Mo (n, γ) ^{93m}Mo (T=6.95 時間)，^{98}Mo (n, γ) ^{99}Mo (T=69.7 時間)，^{100}Mo (n, γ) ^{101}Mo (T=14.6 分)が生成し，それぞれ多くの γ 線を放出する．^{93m}Mo, ^{101}Mo はその半減期から 1 分照射し，3 分後あるいは 30 分後に測定できるはずであるが，この測定時には ^{28}Al, ^{56}Mn によるコンプトンピークの妨害が大きく検出できない．また ^{99}Mo については 1 時間照射後，1 週間後の測定で検出できるはずであるが，この測定時には ^{82}Br, ^{140}La の多数の γ 線による妨害があり，今回は測定できなかった．しかし 1 時間照射後，半減期の長い ^{99}Mo を化学分離し，その 740 keV の γ 線を測定すれば定量できる．

したがって，今回検出できなかった植物の重要な生元素である S, P, Mo は化学分離操作を併用すれば放射化分析による定量が可能となるので，今後これらの元素についても分析方法を検討したい．

謝辞 本研究を行なうにあたって γ 線スペクトルの測定に関し種々御便宜を与えて下さった神戸商船大学原子力研究所の道前正勇教授，三宅 [illegible]助教授に謝意を表するとともに，試料を御提供下さった近畿麻薬取締官事務所の遠藤 勝氏に感謝致します．