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Cesium-137 in Japanese Soil and Rice

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ABSTRACT

Japanese soil and rice collected at 15 localities have been analyzed for cesium-137 since 1959 through 1963. The time trend of rice-levels indicates the minimum in 1960 and 1961 and the maximum in 1963, showing more than threefold value of the minimum. A higher dependency was observed of the level of cesium-137 in rice on the fallout rate than either of the cumulative ground deposit or the N-ammonium acetate extractable cesium-137 in soil. Variation with locality is great and the highest rice-level is observed on the northern Japan Sea side, the lowest in the central and southern parts, and intermediate on the northern Pacific side.

INTRODUCTION

The contribution of cereals to the total dietary intake of cesium-137 in Japan was estimated up to 40% in 1960-1961 and approximately 80% of cereals consumed by Japanese is the polished rice[1]. This is one of the reasons why rice has been taken up as an important monitor material. Another reason lies in the fact that in Japan rice is harvested once a year, in autumn, and consequently, the daily contribution by rice remains constant throughout the year beginning November—December when the consumption of the new crops begins.

The body burden of cesium-137 in a population in the near future would be assessed on the basis of estimates on the level of this radionuclide in the total diet. The first step of estimation should be done on rice which contributes a

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constant intake throughout the year, the second being that on seasonal crops such as leafy vegetables and milk.

In order to find out the factor to determine the level in rice of cesium-137 in relation to the fallout rate, the cumulative ground deposit or other variables, the observed data since 1959 have been analyzed. The results of analyses are summarized in the present report.

MATERIALS AND ANALYTICAL METHODS

A systematic sampling of cereals and soils started in 1957 at National Institute of Agricultural Sciences and all samples were collected in cooperation with members of Governmental and Prefectural Agricultural Experiment Stations throughout the country.

The methods of sample treatment prior to the analysis of cesium-137 were reported previously by one of the present authors (Kodaira, 1964)[2]. The soil samples were collected at each of the flooded rice field from the plowlayer, air-dried and passed through a 2-mm sieve prior to extraction and analysis. The sampling to the depth of 16 cm of paddy soil collects more than 90% of the total cesium-137 which is present to the depth of 26 cm from the surface, as shown in Tab. 1, while in an undisturbed soil profile cesium-137 is concentrated more in the upper 0～2 cm layer.

In general, the rice containing more than 50 pc of cesium-137 per killogram and the total cesium-137 in the soil were analyzed by use of a γ-spectrometric technique described in a previous paper (Yamagata and Iwashima, 1964)[3] The rice containing smaller quantity of cesium-137 and the N-ammonium acetate extract

Table 1. Distribution of ^{137}Cs in soil profile (April 1963, HCl extraction)

Farm soil, Niigata Pref. (Takada)

Depth (cm)	pc/kg dry soil	mc/km^2	Concentration mc/cm/km^2
0～2	169	24 (32%)	12
2～12	61.3	43 (57%)	4.3
12～22	11.0	7.7 (11%)	0.77
Total		74.7 (100%)	3.4

Paddy soil, Ishikawa Pref. (Kanazawa)

Depth (cm)	pc/kg dry soil	mc/km^2	Concentration mc/cm/km^2
0～2	238	29 (26%)	14.5
2～16	88	79 (71%)	5.6
16～26	5.3	3.3 (3%)	0.33
Total		111.3 (100%)	4.3

Undisturbed soil, Tokyo (Nishigahara)

Depth (cm)	pc/kg dry soil	mc/km^2	Concentration mc/cm/km^2
0～ 2	247	20 (73%)	10
2～15	13.4	6 (22%)	0.46
15～25	3.1	1.2 (5%)	0.12
Total		27.2 (100%)	1.1

of soil were analyzed with a radiochemical method followed by β-counting (the dipicrylaminate-chloroplatinate method[4]). The hydrochloric acid extract of soil was analyzed for cesium-137 by the combined ammonium molybdophosphate-dipicrylaminate-chloroplatinate method, since the extract contained a bulk quantity of iron and aluminum[5]. The methods for extraction with hydrochloric acid and N-ammonium acetate of soil are described in Appendices I and II.

TIME TREND OF THE LEVEL OF ^{137}CS IN POLISHED RICE

The levels of cesium-137 in the polished rice from different localities have been determined since 1959; the results are summarized in Tab. 2. The time trend of cesium-137 in polished rice from the entire localities indicates on an average the

Table 2. The levels of ^{137}Cs in polished rice from different localities, 1959-1963 (pc/kg fresh)

Prefecture (locality)	1959	1960	1961	1962	1963
Northern Japan Sea side					
Hokkaido (Sapporo)	133	47	65	154	220
Akita (Akita)		45	39	87	182
Niigata (Takada)	66	25	59	30	158
Ishikawa (Kanazawa)	102	33	39	26	57
Northern Pacific Side					
Iwate (Morioka)	75	40	49	120	195
Miyagi (Sendai)	61	32		74	182
Ibaragi (Mito)			29	43	113
Saitama (Koonosu)	49	38	25	25	76
Tokyo (Tokyo)	56	20	35	32	164
Central-and southern part					
Yamanashi (Kōfu)		28	14	31	28
Miye (Tsu)	66	39	41	14	126
Osaka (Osaka)			22	14	30
Okayama (Okayama)			23	12	38
Tottori (Tottori)	36	27	31	46	101
Fukuoka (Tsukushino)	79		43	45	24
Average for year	72	34	37	50	113

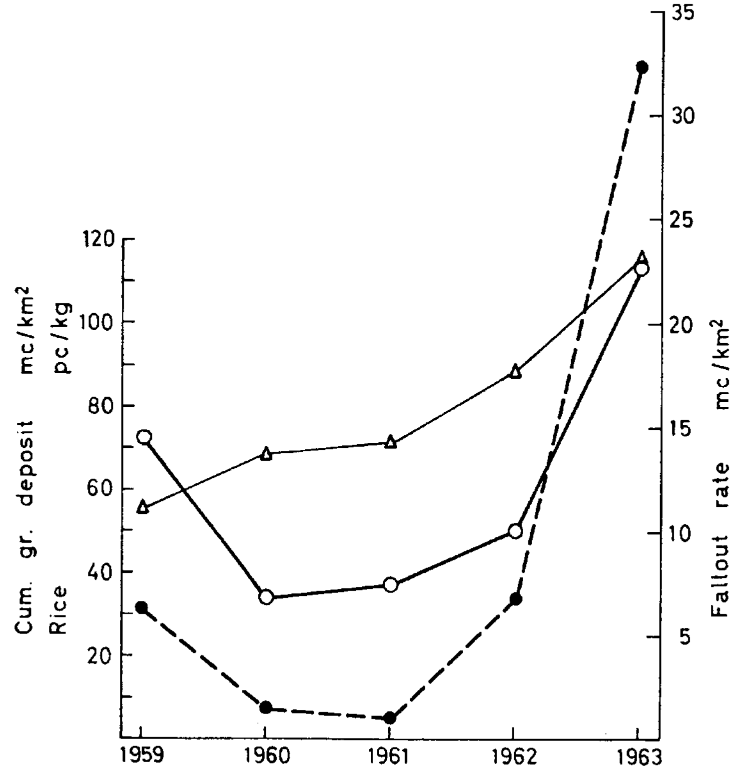

Fig. 1. Time trends of ^{137}Cs in polished rice (average), fallout rate* (June-Sept.) and cumulative ground deposit (up to 31 May of the year)
○ : Polished rice
△ : Cumulative ground deposit
● : Fallout rate
* Measured at the Meteorological Research Institute, Tokyo.

minimum levels in 1960 and 1961 (Fig. 1). The level goes up in the successive years to the maximum in 1963, more than three-fold value of the minumum.

Fig. 1 also shows the time trends of the cumulative ground deposit in Tokyo as summed up to May 31st of the year and the fallout rate as the summation of monthly deposits for June-September. The time trend of the level in polished rice closely resembles that of the fallout rate but not the cumulative ground deposit. This may suggest a higher dependency of the level of cesium-137 in rice to the fallout rate rather than the cumulative ground deposit.

N-AMMONIUM ACETATE EXTRACTABLE ^{137}Cs IN SOIL

1. *Versus cesium-137 in rice*

Root absorption of potassium by plant depends on available potassium in soil and the N-ammonium acetate extraction (pH 7) has been considered one of the pathway through foliar deposition is believed to be more important.

For the purpose of finding, if any, the relationship between deposition of cesium-137 and level of activity in rice, the N-ammonium acetate leachate has been analyzed since 1959 of the soils on which rice crops (shown in Tab. 2) have been raised. The results are summarized in Tab. 3. The plot of the level in rice on the ordinate and the N-ammonium acetate extractable cesium-137 on the abscissa showed no simple linearity. There was also no linearity between the nationwide averages.

The next step of analysis of the relationship was done by grouping geographically the sampling sites in view of agricultural and climatological conditions. A characteristic pattern is known in this country for the fallout deposition rate or radioactivities in some crops, *i.e.,* higher in the northern and Japan Sea side districts and lower in the southern and Pacific side. Climatic conditions also correlate with this pattern; for example, the northern Japan Sea side is characterized by a heavy snow-fall in winter in contrast to dry winter in the Pacific side.

The sampling sites were grouped into three as shown in Tab. 2 and Fig. 2. The plots of the level in rice *vs.* the N-ammonium acetate extractable cesium-137 in soil are shown in Fig. 3 for the northern Pacific side (a) and the northern Japan Sea side (b). In both cases, the lines drawn most fittingly for 1959 or 1963 are best measure of available potassium in soil. By analogy, the N-ammonium acetate extraction has been applied by several workers to the fallout cesium-137 in studying the relation of the uptake by plant and the deposit in soil, although the direct

Table 3. N-ammonium acetate extractable ^{137}Cs in paddy soil from different localities, 1959–1963 (mc/km^2)

Prefecture	1959	1960	1961	1962	1963
Hokkaido	17		10	15	89
Akita	18	25	8	30	51
Niigata	9	14	10	22	53
Ishikawa	24	29	14	26	52
Iwate	2.4	14	12	17	39
Miyagi	13		16	14	17
Ibaragi	30		10	9	7
Saitama	6.4		8	6.6	13
Tokyo	12		9	9	10
Yamanashi	8		3		16
Miye		8	4		29
Osaka			4	4	3
Okayama	2.5		13	6	13
Tottori	13	10	16		23
Fukuoka		12	6	17	20
Average for year	**13**	**16**	**9.5**	**12**	**29**

* Localities are identical to those in Table 2

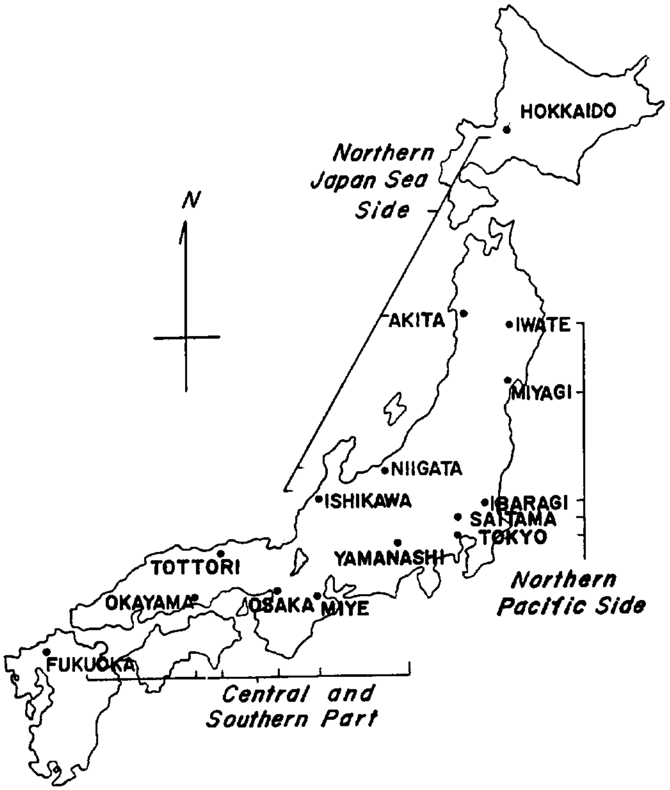

Fig. 2. Sample localities of rice and soil

steeper than those for 1960 or 1961. In view of the higher fallout rates in the former periods than in the latter ones, the above relations may reflect different rate of contribution of the direct foliar contamination in different period if an assumption is made that the amount of cesium-137 in rice consists of two fractions, one of which is proportional to the N-ammonium acetate extractable cesium-137 in soil and the other proportional to direct contamination.

2. *Versus total ^{137}Cs in soil*

Cesium is known to become readily fixed by soil. However, the fixation is not tight enough to become completely unavailable to plants depending on plant species and soil conditions. A part of cesium-137 previously held by soil colloid can be ionically exchanged and liberated by ammonia ion which dominates under reductive condition of paddy soil due to the microbiological oxygen consumption[6)].

Tab. 4 summarizes some of the soil characters at the sampling sites together with the N-ammonium acetate extractable fraction in 1961 and 1963 (autumn) in per cent of the total cesium-137 in soil. It indicates comparatively low percentages of extraction in soils with larger clay content and *vice versa*. This can be reasonably interpreted by the high fixing capacity of clayey minerals.

Another information which can be obtained from Tab. 4 is that in most localities

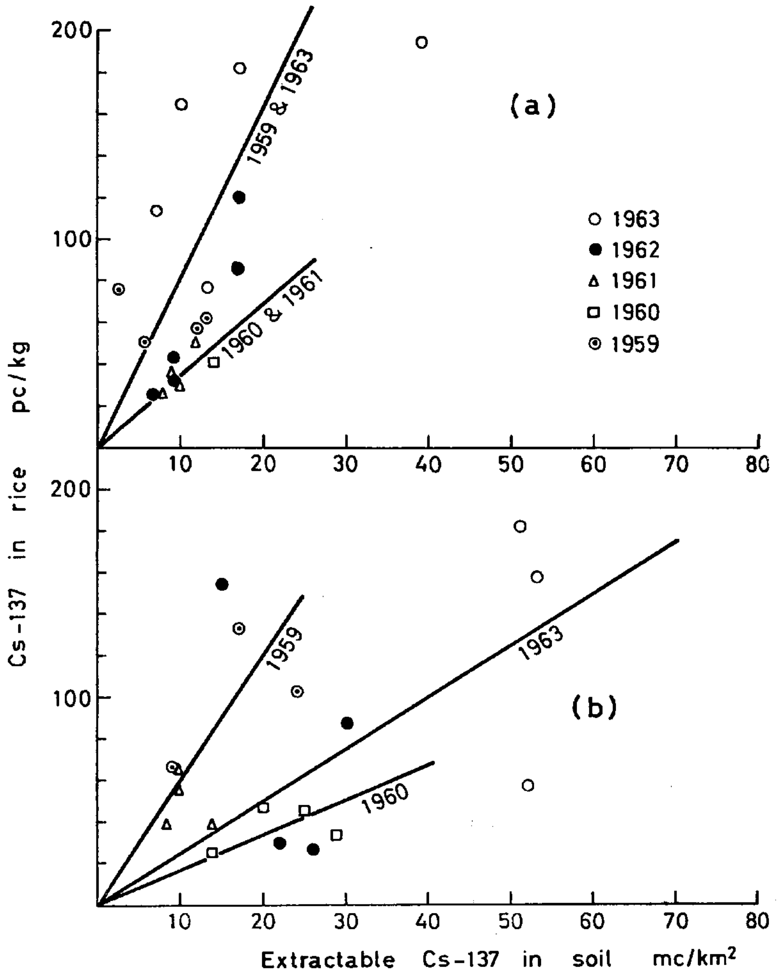

Fig. 3. Relation between the level of ^{137}Cs in the polished rice and N-ammonium acetate extractable fraction in soil
a) Pacific side district b) Northern Japan Sea side

the percentage extraction of cesium-137 is larger in 1963 than in 1961; on the average, it increased from 20.6% in 1961 to 26.9% in 1963. This may be due to the effect of aging on fixation of cesium-137 by soils. As will be discussed in the following section, cesium-137 introduced into soil may become fixed with the passage of time. Therefore, under condition of higher deposition rate, the percentage of extractable fraction becomes larger than in a lower deposition rate and this is the case in 1963 when the rate was 32.3 mc/km^2 (June-September) in contrast to only 1.06 mc/km^2 in 1961 (Fig. 1).

Conclusively, the N-ammonium acetate extractable cesium-137 in soil is not a simple function of the cumulative ground deposit even if the soil is specified, but it is also affected by the doposition rate.

Table 4. N-ammonium acetate extractable ^{137}Cs in paddy soil from different localities (per cent of total ^{137}Cs in soil)

Prefecture	Soil texture	Clay* content	Humus** content	Extractable-^{137}Cs (%) 1961	Extractable-^{137}Cs (%) 1963
Hokkaido	Loam	s	m	27.0	—
Akita	Clay loam	m	s	20.5	30.9
Niigata	Light clay	l	m	12.8	31.6
Ishikawa	Loam	s	s	24.6	44.5
Iwate	Loam	s	l	44.5	37.5
Miyagi	Clay loam	m	s	38.1	34.7
Ibaragi	Caly loam	m	s	33.3	11.7
Saitama	Silt clay	l	m	10.5	16.7
Tokyo	Light clay	l	m	17.3	26.3
Yamanashi	Loam	s	s	4.3	29.1
Miye	Clay loam	m	s	4.7	21.0
Osaka	Light clay	l	s	4.5	8.1
Okayama	Clay loam	m	s	26.0	24.6
Tottori	Clay loam	m	s	—	32.9
Fukuoka	Sandy loam	s	s	—	27.1
		Average for the year		**20.6**	**26.9**

* s: <15%, m: 15~25%, l: 25~45%

** s: 2~5%, m: 5~10%, l: 10~20%

EFFECT OF AGING ON FIXATION OF ^{137}Cs BY SOILS

Tracer experiment was conducted in order to investigate the fixation by soils of carrier-free cesium-137 with the passage of time. Five representative soils consisting of four paddy soils and one field soil were collected in Gunma Prefecture and used for this experiment: No. 1 peaty soil, No. 2 clayey loam, No. 3 clayey loam (Tertiary origin), No. 4 loam and No. 5 alluvial sandy loam.

Carrier-free cesium-137 was diluted with irrigation water and applied to the air-dried soils. Soil samples of 5 g each were taken at intervals after the application and extracted with 10 ml of N-ammonium acetate (pH 7), employing a similar method as described in Appendix 1. The extracts and soils were then γ-assayed. The results are shown in Fig. 4 by plotting the percent extraction of the total cesium-137 in soil on the ordinate and the aging time in a logarithmic scale on the abscissa.

Freshly added cesium-137 became unextractable form in a very short time and a decrease in extractable fraction with the passage of time is exponential in general feature. The extent to which the cesium-137 can be fixed differs in the five soils used in the experiment and there seems some interrelationships between fixation and the fine particle content of the soil. For example, the order of increasing capacity of fixation after one year of aging completely agrees with that of increasing content of particles finer than 70μ, No. 5: 10.5, No. 1: 25.8, No. 4: 31.4, No. 3: 41.5 and No. 2: 48.8%. This phenomenon can be interpreted by the fact that in a mechanical analysis, the fraction finer than 70μ necessarily contains a greater part of clay and the large fixing capacity of some clay minerals.

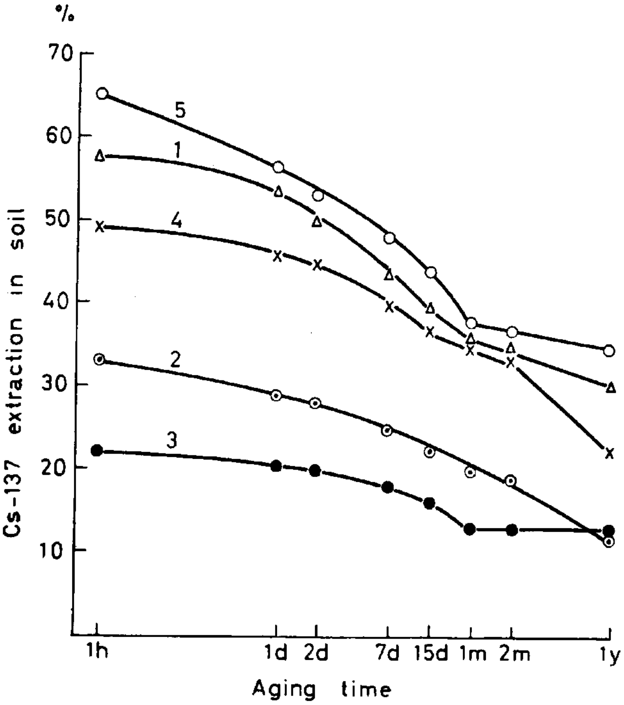

Fig. 4. Effect of aging on fixation of ^{137}Cs by soils (Extraction with N-ammonium acetate, pH 7)

In a very short time, one hour after the application, a considerable fraction of cesium-137 became fixed and only 22~65% remained extractable. The extractable fraction decreased to about a half, 13~38% in one month and after then a gradual decrease occurred, reaching 12~35% in one year. Therefore, in Fig. 4 there is an inflection point in general at the aging time of one month.

Chemical extraction procedure does not necessarily give adequate information on the availability to plants of cesium-137, however, the period of one month may be considered reasonable in which root absorption of freshly added fallout should be taken into consideration to a greater or lesser extent.

EXTRACTION OF SOIL WITH HYDROCHLORIC ACID

In the course of investigation, the extraction with hydrochloric acid was intended in order to find whether a consistent relationship between the total and the extractable cesium-137 in soils was present or not. If such consistency was present, γ-spectrometric determination could be substituted by the extraction and radiochemical determination. The method of extraction is described in Appendix 2.

The results summarized in Table 5 indicate variable percent extraction in different soils, showing the range of 24 to 97%, the average being 60% in 1963 and 38% in 1961. A similar trend as in the N-ammonium acetate extraction is again observed: larger extraction in 1963 than in 1961.

Conclusively, the hydrochloric acid extraction failed in substituting γ-spectrometric method of determining the total cesium-137 in soil because of large variability of percentage extraction among soils.

Table 5. Extraction of paddy soils with hydrochloric acid

Prefecture	1963		1961	
	Total Cs-137 mc/km^2	% Extraction	Total Cs-137 mc/km^2	% Extraction
Hokkaido	—	—	37	43
Akita	165	33	—	—
Niigata	168	36	78	32
Ishikawa	160	73	—	—
Iwate	104	60	27	59
Miyagi	49	63	—	—
Ibaragi	60	52	—	—
Saitama	78	38	76	24
Yamanashi	—	—	70	40
Miye	—	—	86	31
Okayama	53	85	50	36
Tottori	70	97	—	—

Table 6. Local variation of ^{137}Cs in rice in relation to direct contamination and root absorption (1963)

Prefecture	^{137}Cs in rice (pc/kg)		^{137}Cs in soil NH$_4$OAc extractable (mc/km^2)	^{90}Sr fallout* (mc/km^2)	
	In husk	In polished rice		Jun.-Sept. 1963	Cumulative up to Jun. 1963.
Northern Japan Sea side					
Hokkaido	308	220	89	9.29	39.5
Akita	346	182	51	12.33	63.1
Niigata	294	158	53	—	—
Ishikawa	70	57	52	—	—
Av. except Ishikawa	**316**	**187**	**64**		
Northern Pacific side					
Iwate	164	195	39		
Miyagi	101	182	17	9.00	43.7
Ibaragi	330	113	7	—	—
Saitama	283	76	13	—	—
Tokyo	138	146	10	10.38	42.1
Av.	**203**	**146**	**17**		
Central-and southern part					
Yamanashi	142	28	16	—	—
Miye	88	126	29	—	—
Osaka	117	30	3	6.35	28.2
Tottori	276	101	23	—	—
Fukuoka	136	24	20	5.06	47.3
Av.	**140**	**58**	**17**		

* Data obtained from Meteorological Research Institute, Tokyo.

GEOGRAPHICAL VARIATION

In Tab. 6 are shown the fallout rate and deposit of strontium-90 in several sites determined at the Meteorological Research Institute as a reference for local variability in the deposition. The northorn Japan Sea side district is characterized by the higher fallout rate and consequently the larger cumulative ground deposit. Tab. 5 also shows the largest deposition of cesium-137 in this district among those grouped into three. The amount of ammonium acetate extractable cesium-137 in soil is not a single function of the deposition as discussed before but depends on the soil character. The higher availability of cesium-137 in the soils in the northern Japan Sea side district coupled with the higher deposition rate (Tab. 6) brings about the highest contamination of rice.

In contrast, the low availability in the soils in the central- and southern part and the low deposition rate lead to the lowest contamination with only one exception of Tottori Prefecture where the fallout deposition rate is rather high. The northern Pacific side district is intermediate because of the low availability in soil and higher deposition rate.

The geographical variation in the total body burden of cesium-137 in Japanese was assessed by blood analysis and the higher burdens in the northern and Japan Sea side districts and the lower ones in the southern and Pacific side were found7). The good agreement in the pattern of body burden and rice-level suggests the effect of local consumption of foodstuffs.

CONCLUSION

Attempts have been made by several workers to establish quantitative relationships between the levels of activity in foodstuffs, on one hand, and the rate of deposition and the cumulative deposit, on the other, using equations of the type:

$$C = aF_d + bF_r$$

where C represents an activity-level in a foodstuff, F_d the cumulative deposit, F_r the rate of fallout and a and b are proportionality factors.

A lack of available data for local deposition of cesium-137 hampered the nationwide analysis of corelation between the rice-level and deposition and restricted to the consideration of Tokyo district only. Taking the cumulative deposition at the time the end of July and the rate of fallout deposition during the time period June-September, the corelation analysis in Tokyo district has been made.

The result indicated quite a different relative contribution by the two pathways. It suggests that in 1960 and 1961 when the fallout rates were low, 86% of the total cesium-137 incorporated in the polished rice was via the pathway of root absorption, the remainder *via* the aerial absorption. In contrast, in 1963 when the deposition rate was high, 72% was contributed by the direct contamination, the remainder by root absorption.

This finding for 1960 is in good agreement with the result obtained by Ichikawa *et al* who stated the contribution of direct contamination from fallout was 20% of the total cesium-137[8]).

Cesium can be absorbed not only through roots but also through plant body surfaces such as leaves and husks and the enhanced levels of cesium-137 comparing with strontium-90 (roughly 10 times as much)[2]) can be interpreted by much easier translocation in the plant body of the former radionuclide. However, the pathway through root absorption should not be minimized in the case of lowland rice, in which the availability of cesium-137 from the paddy soil is considerable.

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Appendix 1. Method of extraction of ^{137}Cs from soil with ammonium acetate solution

1) Take 400 g of air-dried soil and divide into two portions of 200 g each. Put each portion into a one liter shaking bottle, add 400 m*l* each of ammonium acetate solution (1 M, pH 7) and shake for one hour, then stand overnight.

2) Filter the mixture by suction through a filter paper of loose texture by use of a Buchner funnel and wash the solid with 1.5 liter of the ammonium acetate solution.

3) Add 2 m*l* of cesium chloride carrier solution (Cs^+ 10 mg/m*l*) to the combined filtrate and evaporate the solution to a small syruppy volume.

4) After cooling, cover the container with a watch glass and add 5 m*l* and 20 m*l* of conc. hydrochloric and conc. nitric acids, respectively. Remove the cover and evaporate the solution to dryness.

5) Repeat this treatment until organic materials and/or ammonium salts completely disappear.

6) Moisten the evaporation residue with hydrochloric acid (1 : 1) after cooling, add 50 m*l* of hot water, mix well and then filter. Repeat this treatment several times and discard the insoluble residue. Combine the filtrate and proceed with the analysis of cesium-137 by the combined dipicrylaminate-chloroplatinate method.

Appendix 2. Method of extraction of ^{137}Cs from soil with hydrochloric acid

1) Take 200 g of air-dried soil into a one-liter shaking bottle, add 500 m*l* of hydrochloric acid (1 : 1) and 2 m*l* of the cesium chloride carrier solution, shake mechanically for one hour and then stand overnight.

2) Filter the supernatant through a filter paper of loose texture by use of a Buchner funnel. Wash the residue with about 300 m*l* of 0.1 *N* hydrochloric acid. Combine the filtrate and washing.

3) Take the insoluble solid into the shaking bottle with 250 m*l* of water, add 250 m*l* of conc. hydochloric acid, shake for one hour and then stand overnight.

4) Filter and wash as in step (2) and combine the filtrate and washing with those obtained in step (2).

5) Add 2 g of ammonium molybdophosphate to the combined solution, agitate for 15 minutes and then stand for a while.

6) Discard the supernatant, filter off the solid and wash once with nitric acid (1 : 100).

7) Put the solid with filter paper into a beaker, add sodium hydroxide solution (1 *N*) dropwise with vigorous agitation just to the point ammonium molybdophosphate has dissolved completely.

8) Filter the solution and wash with water completely. Combine the filtrate and washings and proceed with the analysis of cesium-137 by the combined dipicrylaminate-chloroplatinate method.