The Different Distribution of Rubidium and Cesium in Natural Plants

By Noboru YAMAGATA, Toshiko YAMAGATA and Shunji MATSUDA

(Received October 9, 1958)

The uptake of rubidium and cesium by plants in relation to potassium absorption was studied by several investigators. Collander¹⁾ found that the differences between the plant species, the eleven phanerogams studied, were only moderate for potassium, rubidium and cesium contents; all plants absorbed rubidium and cesium with almost the same rapidity as for potassium. The data of Collander and the experiments of other investigators2-4)

suggested the possible use of radioactive rubidium-86 as a tracer for potassium in plant growth experiments, since an isotope of potassium having a suitable half-life was not readily available. On the other hand, cesium-137 is one of the principal long-lived products of nuclear fission and its absorption by cultured plants is a matter of keen interest.

If cesium is hardly differentiated in uptake by plants as in the case of rubidium, and its translocation up to the aerial part is similar to that of rubidium, the distribution of these elements in the leaves of natural plants must be of the same nature. The present paper points

¹⁾ R. Collander, Plant Physiol., 16, 691 (1941).

²⁾ E. Epstein, Proc. Fourth Ann. Oak Ridge Summer Sym., TID-5115, 418 (1952).
3) F. J. Richards, Ann. Bot., 8, 323 (1945).

⁴⁾ J. C. Martin et al., Soil Sci. Soc. Amer. Proc., 10,

TABLE I RUBIDIUM AND CESIUM CONTENT OF NATURAL PLANT (Dry basis)

Name	Location	К %	Rb p.p.m.	Cs p.p.m.
Usnea longissima	Ozegahara, forest	0.249	0.8	0.01
"	","	0.256	0.7	0.004
Thelypteris oligophlebia	Ōmine-mine	2.92	50	0.05
Lepisorus Thumbergianus	Ozegahara, forest	1.07	4	0.1
Dicranopteris glauca	Mt. Õdaigahara	0.631	20	0.01
Osmunda cinnamomea	Ozegahara, bog	1.61	5	0.1
"	// , forest	1.83	6	0.3
Equisetum arvense	Ōmine-mine	5.02	30	0.01
"	"	2.27	20	0.05
Abies Mariesii	Mt. Zaō	0.287	3	0.05
Tsuga Sieboldii	Ozegahara, forest	0.601	2	0.03
Pinus densiflora	Tsubaki, sea-shore	0.378	2	0.01
P. pumira	Mt. Zaō	0.346	3	0.05
P. parvifolia	Mt. Shibutsu	0.347	1	0.01
Myrica gale var. tomentosa	Ozegahara, bog	0.354	2	0.08
Ostrya Japonica	Mt. Zaō	0.802	10	0.02
Fagus japonica	"	0.514	10	0.05
Quercus crispula	Mt. Ōdaigahara	0.816	5	0.02
"	Ozegahara, forest	0.707	2	0.4
"	Mt. Shibutsu	0.531	2	0.01
Reynoutria japonica	Mt. Shibutsu	0.837	3	0.6
Dianthus superbus var.	Mt. Shibutsu	0.964	4	0.3
Nuphar pumilum var. ozeense	Ozegahara, bog	0.887	10	0.05
Nymphaea tetragona	","	0.743	10	0.05
Ranunculus nipponicus var.	", "	1.91	5	0.3
Magnolia obovata	Mt. Zaō	0.866	10	0.02
"	Ōmine-mine	2.01	20	0.004
Hydrangea hirta	"	1.22	10	0.3
Saxifraga Fortunei	Mt. Shibutsu	0.801	1	0.01
Sorbus commixta	"	0.987	4	0.002
Sanguisorba sitchensis	"	1.15	4	0.03
Sieversia pentapetala	"_	0.310	2	0.05
Skimmia japonica	Mt. Ōdaigahara	0.726	6	0.2
Empetrum nigrum var.				
japonicum	Mt. Shibutsu	0.233	1	0.05
Acer Tschonoskii	Mt. Zaō	0.677	6	0.05
A. Mono	Ozegahara, forest	1.39	4	0.05
Aesculus turbinata	","	1.20	3	0.02
Tilia japonica	","	1.71	5	0.03
Kalopanax pictus	", "	1.90	6	0.07
Phyllodoce aleutica	Mt. Zaō	0.338	3	0.02
Rhododendron sp.	"	0.677	6	0.05
R. Tchonoskii	Mt. Shibutsu	0.324	0.8	0.05
R. Fauriae	"	0.300	1	0.05
Andromeda Polifolia var.				
grandiflora	Ozegahara, bog	0.456	1	0.05
Arcterica nana	Mt. Shibutsu	0.173	0.6	0.1
Menyanthes trifoliata	Ozegahara, bog	1.81	6	0.4
Adenophora nikoensis	Mt. Shibutsu	2.64	8	1
,,	//	2.10	3	0.1
Cirsium spicatum	Mt. Zaō	1.25	30	0.3
,	Omine-mine	1.90	20	0.04
Sasa paniculata	Mt. Ōdaigahara	0.490	4	0.01
S. oseana	Ozegahara, bog	1.34	8	0.1

Name	Location	К %	Rb p.p.m.	Cs p.p.m.
Miscanthus sinensis	Gunma-mine	0.171	1	0.1
"	Mt. Zaō	0.934	30	0.2
"	Ozegahara, bog	0.869	20	1
Phragmites communis	", "	1.37	4	0.5
Lisichiton camtschatcense	", "	6.45	25	0.5
Veratrum stamineum	", "	4.35	30	0.01
"	Mt. Shibutsu	2.71	8	0.05
Narthecium asiaticum	Ozegahara, bog	1.98	10	0.1
Iris laevigata	" . "	1.63	4	0.05

out the different distribution of rubidium and cesium in natural plants and deals with the discussion of the different nature of these elements in the biological cycle from soil to plant.

Distribution of Rubidium and Cesium in Natural Plants.—The distribution studies on the rare alkali elements in soil, plants, and waters have been carried out and reported by one of the authors⁵⁾. Sixtyone samples of natural plants consisting of fifty different species were collected from various districts in Japan analyzed for leaves by a spectrographic method by use of cathode-layer arc excita-The plant species consist of two mosses, seven ferns, five gymnosperms, and eleven monocotyledons, the rest being dicotyledons. The analytical data are summarized in Table I. Rubidium varied from 0.6 to 50 p.p.m. and cesium from 0.002 to 1 p.p.m. on the dried material.

The determination of rubidium and cesium in plants has been reported by G. Bertrand and D. Bertrand⁶⁾. The values for land plants examined by them ranged from 2.1 to 81 (average, 20.3) p.p.m. of rubidium and from 3 to 88.5 (average, 22) p.p.m. of cesium for phanerogams on the dried material. The values for rubidium are in good agreement with those given by the authors; however, the values for cesium given by Bertrand et al. are an order similar to the values for rubidium. showing surprisingly high values. Recently, Smales and Salmon analysed seaweed and the ratios of $0.55 \sim 1.4 \times 10^{-2}$ (average, 1.0×10^{-2}) were given for the ratio Cs/Rb⁷⁾. Rubidium is more abundant than cesium by factors of approximately 100 in rocks, soils, seaweeds, and other natural materials; the land plants could not be the only

exception. The result of the authors' determination indicates the variation of the Cs/Rb ratio from 2×10^{-4} to 2×10^{-1} and the average 3×10^{-2} .

Relationships between the potassium content and the rubidium or the cesium content of natural plants were examined;

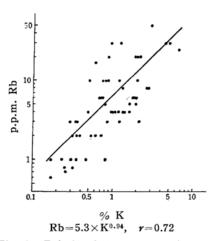


Fig. 1. Relation between potassium and rubidium contents of natural plant.

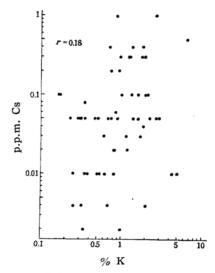


Fig. 2. Relation between potassium and cesium contents of natural plant.

N. Yamagata, J. Chem. Soc. Japan, Pure Chem. Sec. (Nippon Kagaku Zasshi), 71, 288, 567 (1950); 72, 154, 157, 247, 299, 530, 610, 753 (1951); N. Yamagata and T. Kurobe, ibid., 72, 944 (1951).

G. Bertrand and D. Bertrand, Compt. rend., 219, 325 (1944); 229, 453 (1949).

⁷⁾ A. A. Smales and L. Salmon, Analyst, 80, 37 (1955).

TABLE II
RUBIDIUM AND CESIUM CONTENTS OF SOIL IN JAPAN
(Dry basis, hot conc. HCl soluble)

Soil Type Location		К %	Rb p.p.m.	Cs p.p.m.
Kwantō loam	Meguro, Tokyo	0.04	0.8	$0.02 \sim 0.2$
// , uppon layer	Nishigahara, Tokyo	0.05	0.8	$0.1 \sim 1$
// , lower layer	" "	0.04	0.6	$0.01 \sim 0.1$
Volcanic ashy soil	Hokkaido	0.02	0.3	$0.06 \sim 0.6$
Bog soil, upper layer	Aomori Pref.	0.13	20	0.5
Meadow soil, upper layer	Akita Pref.	0.08	8	$0.1 \sim 1$
", middle layer	"	0.07	40	0.4
// , lower layer	"	0.05	10	0.2
Volcanic ashy soil	Ibaragi Pref.	0.01	2	0.03
Tertiary diluvium, upper layer	Aichi Pref.	0.04	3	$0.03 \sim 0.3$
" lower layer	"	0.06	10	0.2
Alluvial paddy soil	Shiga Pref.	0.11	80	0.3 ~3
Granitic alluvium	Shiga Pref.	0.07	20	$0.2 \sim 2$
Sandy alluvium (I)	Kagawa Pref.	0.07	10	0.03
// (II)	"	0.07	2	$0.04 \sim 0.4$
Average		0.06	14	(0.3)

the Rb/K ratio varied from 1.2×10^{-4} to 3.2×10^{-3} and the average was 7.3×10^{-4} for sixty-one samples. The variation of Cs/K ratio was greater than that of Rb/K and covered from 2×10^{-6} to 1×10^{-3} . Fig. 1 is a plot in logarithmic scale of the potassium content in percent versus the rubidium content in p.p.m. The existence of a linear relationship between rubidium and potassium contents can easily be seen; the coefficient of correlation (r) is 0.72. Fig. 2 is a plot in logarithmic scale of the potassium versus the cesium content. In this case, the existence of a linear relationship is doubtful (r=0.18). The coefficient of correlation between rubidium and cesium contents was also calculated and the value of 0.0073 appears to indicate no relation between the contents of these elements in the examined plants.

The variation of the Cs/K ratio greater than the Rb/K ratio and the doubtful existence of correlation between cesium and potassium contents may be accounted for by one or several reasons as under described.

- (1) The variation of total cesium content of the soils on which the plants were growing is greater than that of rubidium.
- (2) The uptake of cesium into the aerial portions of the plants is irregular as compared with that of rubidium; for example, the uptake tends to increase at the higher concentrations of cesium in soil solution.
- (3) The ion-exchange or adsorption mechanism between soil and solution that makes the release of cesium and rubidium for soil colloids and minerals is different.

Distribution of Rubidium and Cesium in Soil.—Soils usually contain 20~5008 or 5~ 100 (average, 60^{9}) or 100^{10}) p.p.m. of total rubidium and only less than 1 to 25.7 (max.) p.p.m.8) of total cesium in contrast with 1.36% of total potassium on the average⁹⁾. Although the data for cesium are too incomplete to guess the average content, Vinogradov gave a value of approximately 5 p.p.m.9,10). The average ratio of Rb/K is $4\sim7\times10^{-3}$ and the average Cs/K is approximately 4×10^{-4} . data appear to be reasonable in accordance with Horstman's recent work11). He gave 240~460 for the average ratios K/Rb of various rocks (Rb/K= $2.2\sim4.2\times10^{-3}$) and 6700 for K/Cs of granitic rocks (Cs/K= 1.5×10^{-4}). On the weathering of rocks, absolute loss of the three elements occurs with the fastest leaching and the least adsorption of potassium, as the result, the ratios of Rb/K and Cs/K increase. However, the above-mentioned data indicate that soils differ not so much from fresh rocks in the averaged ratios of Rb/K and Cs/K.

No biological activities can ever extend to the complete decomposition of soil forming silicate minerals, although the total elements existing in soil are reserved

⁸⁾ D. J. Swaine, "Trace-Element Content of Soils". Commonwealth Bureau of Soil Sci. Tech. Comm., No 48 (1955) p. 34, 88.

⁹⁾ V. I. Vernadsky, "Selected Papers" (in Russian), ed. by A. P. Vinogradov, Acad. Sci. U. S. S. R. Moscow (1954) Vol. I, p. 365.

⁽¹⁹⁵⁴⁾ Vol. 1, p. 505.

10) A. P. Vinogradov, "Geochemistry of Rare and Disperse Chemical Elements in Soil" (in Russian), Acad. Sci. U. S. S. R. Moscow (1957) p. 86-94, 216.

11) E. L. Horstman, Geochim. et. Cosmochim. Acta, 12, 1-28 (1957).

for living matter to be utilized in far future. In this respect, we often consider the amount of elements extractable with mineral acid as a measure for the availability by plants in the near future.

Fifteen representative Japanese soils were extracted with hot hydrochloric acid (31%) and analyzed; the result¹² has been recalculated and summarized in Table II. The variation of rubidium and cesium covered from 0.3 to 80 and from 0.01 to 3 p.p.m. respectively when potassium varied from 0.01 to 0.13%. The Rb/K ratio varied from 1.5×10^{-3} to 7×10^{-2} and the Cs/K ratio varied from 4×10^{-5} to 1×10^{-3} . These values indicate that the variations of levels of rubidium and cesium in soil are of the same order and it appears to us that the first reason (1) for the different distribution of these elements in natural plants fails to elucidate the

Culture Experiment.—Many investigators carried out the culture experiments with solutions of various concentrations of rubidium and cesium; an example is shown in Table III. When the uptake by natural or cultivated plants growing in soil comes into question, we must consider the natural levels of rubidium and cesium concentrations in soil. Indeed, the culture experiment with higher concentrations may serve for certain physiological purposes, however, for the present purpose, it will be desirable to make the levels of these elements in culture solution as close as possible to those in natural soil conditions.

TABLE III
RUBIDIUM, CESIUM AND POTASSIUM LEVELS
IN CULTURE SOLUTION USED BY SEVERAL
INVESTIGATORS

	Rb meq./1.	Cs meq./l.	K meq./l.
Collander ¹⁾	0.1	0.1	2
Menzel and Heald ¹³	0.054 0.0054	$0.054 \\ 0.0054$	0.54
Murphy et al.14)	Rb/K (v	weight)	

It is not the total or acid soluble but only a fraction of the elements that the root of a plant can readily absorb. It is well known that rubidium and cesium are more firmly held by the clay fraction of soil than potassium, probably on account of their greater ionic polarizability. We can therefore assume that the Rb/K and the Cs/K ratios in the neutral ammonium acetate extract of soil or in the actual soil solution are probably smaller respectively than the ratios of the total or strong acid soluble elements in soil.

As the concentrations of available or active rubidium and cesium in soil had not been estimated when the present cultue experiment was undertaken, the standard ratios of Rb/K and Cs/K were temporarily fixed to approximately 10^{-2} and 2×10^{-4} respectively except where varied as an experimental condition.

Experimental

The experiment was conducted in a green-house and cucumber, rice-plant, and soy-bean were chosen for the experiment plants. The seedlings were grown in nutrient solutions in 3 liter porcelain pots from 3rd July to 11th July when tracer isotopes were added. After that, a long period experiment was carried on for thirty days with occasional renewal of nutrient solution and radio isotopes.

The culture solution was prepared with extra pure chemicals and distilled water; the elementary composition was as follows¹⁵): (p.p.m. in solution)

NO₃-N: 130, NH₄-N: 23, K: 200, Ca: 200, Na: 20, Mg: 40, Si: 20, P: 50, Cl: 20, S: 50, Fe: 2, Mn: 0.4, B: 0.4, Zn: 0.4, Cu: 0.04, Mo: 0.002.

Two isotopes ¹³⁷Cs and ⁸⁶Rb were added together in a concentration of about $10 \,\mu\text{c/l}$. each of nutrient solution. Another experiment to determine the uptake velocity of these elements by plants in a short period was conducted with higher concentrations of isotopes.

The harvested plant leaves were incinerated and counted for the total activity of 137Cs and After thirty-nine days the sample was recounted for the activity. As the difference was to be ascribed to the decay of a short-lived isotope 86Rb (half-life: 19.5 days), each of the activities of these isotopes could be calculated. After the activity determination, the plant ash was analyzed for potassium by a chemical method. The ratio of counts from 187Cs (or 86Rb) and concentration of potassium in the plant were compared with the same ratio in the culture solution. The distribution factors between plant and solution were calculated by dividing the two ratios: (86Rb/K in plant)/(86Rb/K in solution) for rubidium, and (187Cs/K in plant)/(137Cs/K in solution) for cesium.

N. Yamagata, J. Chem. Soc. Japan, Pure Chem. Sec. (Nippon Kagaku Zasshi), 71, 567 (1950).
 R. G. Menzel and W. R. Heald, Soil Sci., 80, 287

 ¹³⁾ R. G. Menzel and W. R. Heald, Soil Set., 80, 251 (1955).
 14) W. S. Murphy, A. H. Hunter and P. F. Pratt, Soil

¹⁴⁾ W. S. Murphy, A. H. Hunter and P. F. Pratt, Soil Sci. Soc. Amer. Proc., 19, 433 (1955).

¹⁵⁾ N. Yamagata and T. Yamagata, Culture experiment with a universal solution. Presented at the Annual Meeting of the Society of the Science of Soil and Manure, Tokyo, Apr., 1958.

Results

A short period experiment was performed to examine whether there were any differences between rubidium and cesium of the transport velocity into the aerial part. The result of the experiment is shown for rice-plant (Fig. 3). transport velocity is represented by plotting the increase of distribution factors against time, consequently, it does represent an increase of rubidium or cesium in the plant not absolutely but relatively to potassium. Rapid absorption is observed in several early hours and steeper slope for rubidium appears to indicate faster transport in the plant or rubidium than of cesium at the early stage of absorption, although the difference between these elements will have no connection with the absorption during the prolonged period.

A long period experiment was conducted with the three crop plants over a range of concentrations of rubidium and cesium from 0.02 to 20 and about 0.0001 to 4 p.p.m. respectively. The results are shown in The effect of concentration Table IV. was not clear except for rubidium in the case of soy-bean. The relatively small variation of distribution factors with concentration in nutrient solution indicates that the effect of concentration on rubidium and cesium uptake relative to potassium is small, by a factor of two or three at most. The results also indicate that a difference exists between plant species. It can be seen that the distribution factor for cesium is greater in leaves than roots only in the case of soy-bean. plant accumulates less cesium relative to potassium than the other plants. These results are in good agreement with Rediske

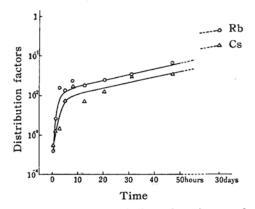


Fig. 3. Uptake velocity of cesium and rubidium by rice-plant as represented by distribution factor between solution and plant leaf.

and Selders' experiement with several other crop plants¹⁶).

Distribution of Rubidium and Cesium in Tobacco Leaf.—The analytical data for the distribution of the alkali elements in cultivated tobacco leaves were previously presented by one of the authors¹⁷⁾. The recorded value for rubidium varied from 5 to 90 p.p.m. on the dried material and the average was 30 p.p.m. The Rb/K ratio varied from 5×10^{-4} to 3×10^{-3} and the average was 1×10^{-3} ; these values are in good agreement with those of the natural plants.

On the other hand, the concentration of cesium in tobacco leaves was very low and for this reason it was only successfully determined quantitatively for one sample among seventeen (No. 10a in the original paper). This sample contained 0.17 p.p.m. cesium in ash or 0.02 p.p.m. in the dried material, and the Cs/K ratio for the whole leaf was 1.2×10^{-6} . The Cs/K ratios for the rest of the sample are considered to be less than this value. It can thus be concluded that the Cs/K ratios for cultivated tobacco leaves are surprisingly low as compared with those for the natural plants $(2 \times 10^{-6} \sim 1 \times 10^{-3})$.

According to the experiment conducted by Menzel¹⁸⁾, the uptake of rubidium and cesium by plants was inversely proportional to the available potassium in the soil, and the uptake of cesium was more affected than that of rubidium by the increase in available potassium. Thus, the remarkable discrimination in the cultivated tobacco plant of cesium from potassium will be accounted for by the possible existence of a greater quantity of available potassium in the soil as compared with the case of natural plants.

Another interesting phenomenon should be pointed out, that of the different distribution with in the tobacco leaf of cesium from rubidium and potassium. The tobacco leaf was divided into two portions, the principal vein and the remaining part, and separately analyzed. Qualitative examination indicated in every sample of tobacco leaves the considerable enrichment of potassium and rubidium in the vein and the inverse trend for cesium. Quantitative determinaion was made with the tobacco leaf No. 10a and the result is

¹⁶⁾ J. H. Rediske and A. A. Selders, U. S. Atomic Energy Comm., HW-35174 (1953).

N. Yamagata, J. Chem. Soc. Japan, Pure Chem.
 Sec. (Nippon Kagaku Zasshi), 71, 288 (1950).
 R. G. Menzel, Soil Sci., 77, 419 (1954).

TABLE IV

DISTRIBUTION FACTORS FOR Rb-K AND FOR Cs-K IN LEAF AND ROOT OF
PLANTS OF SEVERAL CROP SPECIES
(K: 200 p.p.m. in nutrient solution)

Ratios in Nutrient Distribution Factors soln. Pot No. Part Cucumber Rice-plant Soy-bean Rb/K Cs/K (in weight) Rb/K Cs/K Rb/K Cs/K Rb/K Cs/K 1 10-4 5×10^{-7} 0.30 0.66 0.35 0.31 0.17 0.40 R 0.14 0.62 0.26 0.33 0.72 0.28 2 10^{-3} 2×10^{-5} 0.56 0.91 0.57 0.23 0.24 0.82 R0.13 1.75 0.50 0.49 0.65 0.40 3 10^{-2} 2×10-4 0.32 0.40 0.45 0.18 0.39 0.54 R (standard) 0.180.810.45 0.460.65 0.33 L 4 10-1 2×10^{-3} 0.50 0.52 0.88 0.57 0.480.57 R 0.30 1.25 0.32 0.25 0.42 0.30 5 10^{-2} 2×10^{-2} L 0.72 0.570.17 0.150.72 0.72R0.35 0.760.22 0.58 0.24 0.33 L 0.48 0.48 0.61 Average 0.61 0.29 0.40 R0.22 1.04 0.35 0.42 0.54 0.33

Table V

The different distribution in tobacco leaf of cesium from rubidium and potassium (Sample No. 10a, recalculated from the previous report¹⁷⁾)

	K %	Rb p.p.m.	Cs p.p.m.	Rb/K	Cs/K
Whole leaf	1.77	40	0.02	2×10^{-3}	1.2×10^{-6}
Principal vein	3.70	90	0.006	2.5×10^{-3}	1.5×10^{-7}
The remaining part	1.35	20	0.02	1.5×10^{-3}	1.5×10^{-6}

shown in Table V. The Rb/K ratio is similar in the principal vein and in the rest. On the other hand, the Cs/K ratio in the former is approximately one tenth of the ratio in the latter, that is to say, the cesium is relatively enriched in the latter as compared with potassium and rubidium. These results appear to be inconsistent with the conclusions by Glueckauf¹⁹) who considered the possible enrichment of cesium and strontium occurring in flowers, leaf veins and root substrate.

Conclusion

1. The greater variation of cesium in natural plants than that of rubidium could not be attributed to the variations of total or acid soluble elements in the soil.

2. The relative uptake by plants of rubidium and cesium to potassium was only slightly affected by the concentrations of these elements in nutrient solution. It appears that a difference does exist between plant species in their ability to accumulate cesium and rubidium relative to potassium.

3. The greater variation of the Cs/K

ratio and the doubtful existence of correlation between cesium and potassium contents of natural plants may be principally attributed to the complicated mechanisms of ion-exchange or adsorption between soil and solution.

Fixation by soils and uptake by plants of cesium have been investigated by several authors²⁰⁻²²⁾ in connection with the problems of radioactive contamination from nuclear fission explosions and also with a view to the ultimate disposal of fission products. The fate of cesium-137 in the biogeochemical cycle from soil to plant has usually been considered likely to follow that of potassium. This appears not to be true after both the results ascertained by us and other investigators. consideration is taken of the difference of the ionic radii between rubidium (1.49Å) and cesium (1.63Å), we can not expect that cesium can replace potassium (1.33A) as freely as rubidium does. In this regard, the concept of the "Cesium Unit" temporarily adopted by U.N. authorities to evaluate the concentration of cesium-137 in natural

¹⁹⁾ E. Glueckauf, Atomic Energy Research Establishment, Harwell, SPAR/7 (1956).

C. B. Amphlett, Research, 8, 335 (1955).

C. B. Amphlett and L. A. McDonald, J. Inorg. & Nuclear Chem., 2, 403 (1956).

²²⁾ H. Nishita et al., Soil Sci., 81, 317 (1956).

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materials relatively to potassium has little meaning. We believe that the future burden of mankind with radioactive cesium-137 will properly be estimated on the basis

of the concept of isotopic dilution.

Kiriu College of Technology Gunma University, Kiriu