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Memorandum

To: (1) Dr. Walter S. Snyder ✓ Oct. 7, 1974
(2) Dr. Philip Gustafson

From: G. Allan Lough

Subject: Copy of paper by Takizawa and Sugai

Please refer to my letter of September 26, 1974 to Dr. Snyder.

Enclosed herewith is a xerox copy of the Japanese paper which was under discussion on September 19, 1974 at Argonne.

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Plutonium 239, Strontium 90, and Cesium 137

Concentrations in Human Organs of the Japanese

Yukio Takizawa, MD, and Ryuichi Sugai, MD, Niigata, Japan

This is a report of the dynamic aspects of the selective accumulation of radioactive nucleides, such as plutonium 239, cesium 137, and strontium 90. The survey proceeding the Japanese atomic explosion has revealed considerable increase in the amount of fallout. The possible hazards and the need to evaluate the contamination are considered.

RADIOACTIVE contamination of the environment caused by nuclear tests is now creating serious hazards to mankind.[1] Effects of radiation have been studied in detail with reference to the contamination of air, earth, and water, with the final purpose of establishing the degree of environmental contamination that is harmful to human beings. The most probable contamination of humans occurs through food and water.

Generally, the effective dose is very complex in nature depending upon long- and short-lived fission products. In addition, the relationship between the dose and biological effects is also complex, since the effect may develop rather late, taking the form of specific neoplastic diseases, shortened life-span, and genetic abnormalities. The scientific evaluation of the hazard of radiation to human beings is, therefore, quite difficult.

In any attempt to resolve this difficulty, it is of primary importance to know the sources of radiation, the nature of the nucleides, and the distribution throughout various organs of the body.[2]

In our study, the accumulations of radioactive plutonium (^{239}Pu), radioactive cesium (^{137}Cs), and radioactive strontium (^{90}Sr) in human tissues over the period of the last ten years since 1960 were measure for comparison with the possible fluctuations of environmental pollution. Our paper is a report of the survey made thus far.

Materials and Methods

The material selected for study consisted of brain, lung, liver, kidney, spleen, gonads, bones, and muscles from cadavers obtained from the Hospital of Niigata University School of Medicine in northern Japan.

The analytical procedure for measuring ^{239}Pu is illustrated in Fig 1.[3] Each organ was weighed and ashed by a wet method using nitric acid, and the ash was dried over a hot plate. Separation of ^{239}Pu was accomplished through ion-exchange resin chromatography, and the rate of recovery (maximum value 96%, average value, 90%) was calculated after part of the standard solution of ^{239}Pu was added to the sample. Plutonium 239 was used as the sample after

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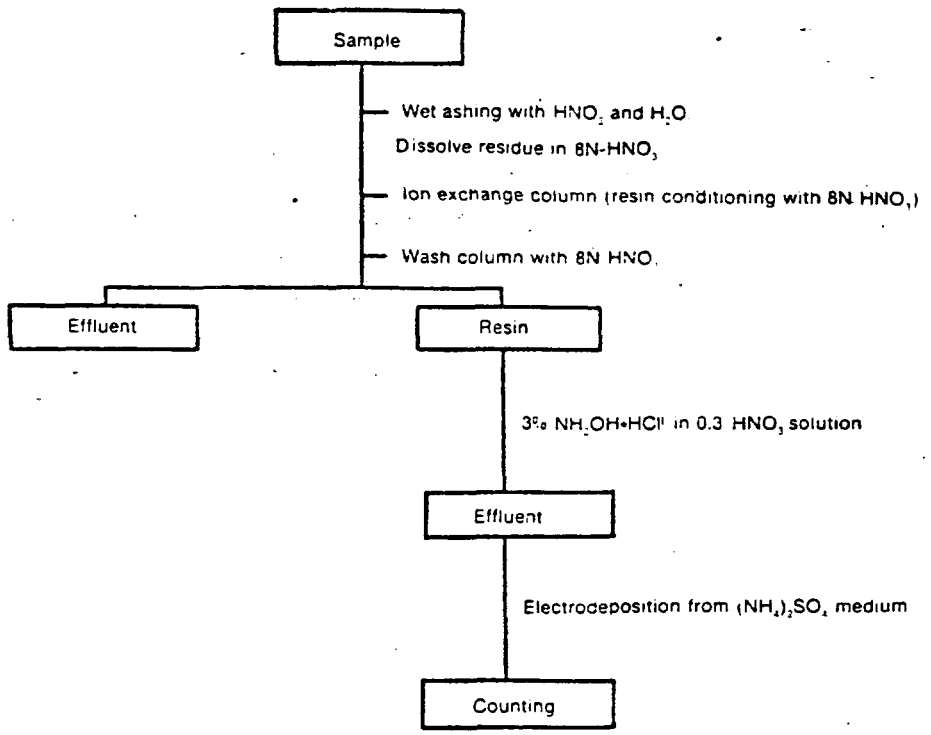

Fig 1.—Analytical procedure of ^{239}Pu in human tissue.

electrochemical displacement on a stainless plate.

Using ammonium phosphomolybdate and dipicrylamine methods, ^{137}Cs was separated and then measured with a low background gas flow counter. Strontium 90 was separated by the fuming nitric acid method. The amounts of ^{137}Cs and ^{90}Sr were determined by the method given in *Analytical Method for Radioactive Cesium and Strontium* published by the Science and Technology Agency of Japan in 1963.

Results and Comment

Since 1954, after the nuclear bomb explosions in Hiroshima and Nagasaki, we have been collecting data to arrive at an estimate of deposited radioactive fallout in Niigata City, Japan.[4-6] The recent gross estimation of radioactivity (counts per minute) in the human body is shown in Table 1. The result shows a decrease to 1/6, 1/40 of that noted around 1957, reflecting fluctuation of the fallout in recent years.[7]

The most important point, however, is the evaluation of cumulative amounts of the long-lived nuclides, ^{90}Sr, ^{137}Cs, and ^{239}Pu. Since the reopening of nuclear testing in 1961, the survey of the Japan Atomic Energy Commission has focused on the amount of ^{90}Sr found in human bone and that of ^{137}Cs found in urine. The accumulation of ^{239}Pu has become a world-wide concern in recent years.

The cumulative amounts of ^{239}Pu found in the principal organs, as is evident from Table 2, was highest in bone, followed by the liver and spleen. The yearly variation of the amount of ^{239}Pu in the lung is shown in Table 3, which indicates some recent increase. The amount of ^{239}Pu in the lung gradually increases until 1965, and reaches a peak value in 1968. A similar trend was seen in bone. Thus, the amount of ^{239}Pu in human bone in Japan was ten times higher than that in the United States as reported in 1965. The obvious increase in environmental contamination is attributed to nuclear tests. The cumulative fallout is already

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Table 1.— *Summary of β-Radioactivity in Human Organs, 1962 to 1966 (Counts per Minute)*

Organs		1962	1963	1964	1965	1966	Overall Mean
Lung	n	8	20	14	15	6	
	Mean	0.41	0.43	0.21	0.13	0.10	0.25 ± 0.05
Liver	n	10	17	11	15	7	
	Mean	0.28	0.25	0.23	0.12	0.07	0.19 ± 0.02
Kidney	n	6	16	13	13	7	
	Mean	0.34	0.29	0.31	0.14	0.14	0.24 ± 0.04
Spleen	n	9	21	15	16	6	
	Mean	0.19	0.22	0.25	0.24	0.22	0.22 ± 0.02
Genital	n	9	29	11	19	6	
	Mean	0.21	0.27	0.32	0.33	0.26	0.28 ± 0.01
Bone	n	7	17	17	19	4	
	Mean	0.18	0.18	0.27	0.26	0.31	0.24 ± 0.03
Muscle	n			18	15	7	
	Mean	....	....	0.14	0.11	0.09	0.11 ± 0.01

Table 2.—*Plutonium 239 in Human Organs, 1963 to 1968 (Curie × 10^{-15}/Wet gm)*

	1963	1964	1968	1069		
	(M) 46 yr	(F) 57 yr	(F) 62 yr	(F) 70 yr	(F) 59 yr	(M) 54 yr
Cerebrum	0.05	.....	0.03	0.10	.....	.....
Lung	.....	0.02	0.19	0.13	0.10	0.26
Liver	0.12	0.12	0.63	0.03	0.05	0.05
Kidney	0.19	0.05	0.09	0.20	0.11	.....
Spleen	0.04	.....	0.22	.....	0.38	0.26
Genital	0.29	0.05	0.31	2.36	2.56	.....
Bone	0.05	0.37	0.64	6.30	0.66	1.78
Muscle	.....	.....	.....	1.80	2.90	1.50

Table 3.—*Plutonium 239 in Human Lung, 1960 to 1969 (Curie × 10^{-15}/Wet gm)*

	^{239}Pu		^{238}Pu	
	n	Mean	n	Mean
1960	5	0.013*	5	ndt
1961	5	0.015*	5	ndt
1964	2	0.012*	2	ndt
	3	0.032*	3	ndt
1965	3	0.038*	3	0.084
	3	0.029*	3	0.020
1966	3	0.031	...	....
1967	3	0.103	...	....
1968	3	0.193	...	....
1969	3	0.167	...	....

* Value in composite sample.
† nd = Not detected.

Table 4.—*Strontium 90 in Human Bones (Ribs), 1962 to 1968 (Micromicrocurie/gm Ca)*

Year	No. Cases	Av Age	^{90}Sr	Range
1962	32	35.2	0.62 ± 0.41	1.47 ~ 0.01
1963	33	38.1	0.64 ± 0.08	1.38 ~ 0.16
1964	26	37.5	0.60 ± 0.11	1.40 ~ 0.07
1965	25	39.7	0.65 ± 0.11	1.53 ~ 0.07
1966	21	37.9	0.85 ± 0.09	1.63 ~ 0.17
1967	15	41.1	1.07 ± 0.45	1.82 ~ 0.57
1968	19	37.0	0.99 ± 0.13	2.91 ~ 0.42

Table 5.—*Cesium 137 in Human Organs, 1962 to 1966 (Micromicrocurie/Wet gm)*

Organs		1962	1963	1964	1965	1966	Overall Mean
Lung	n	5	7	8	4	3	
	Mean	0.06	0.14	0.14	0.14	0.13	0.12 ± 0.012
Liver	n	8	7	6	4	3	
	Mean	0.08	0.10	0.11	0.09	0.10	0.10 ± 0.003
Kidney	n	2	6	4	4	3	
	Mean	0.05	0.15	0.13	0.09	0.09	0.10 ± 0.003
Spleen	n	6	8	6	9	2	
	Mean	0.07	0.11	0.13	0.12	0.14	0.11 ± 0.002
Genital	n	3	7	1	3	2	
	Mean	0.12	0.16	0.28	0.15	0.14	0.17 ± 0.004
Bone	n	4	3	10	12	3	
	Mean	0.18	0.27	0.24	0.27	0.26	0.24 ± 0.012
Muscle	n	3		3	9	3	
	Mean	0.07	.....	0.18	0.08	0.12	0.11 ± 0.007

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It is of interest to note that ^{238}Pu was detected only in the lung sample obtained in 1965. As an example of such fallout, the accident of the artificial satellite (SNAP-9A) hich burned in the stratosphere over the Indian Ocean in 1964 may be considered.[2,3]

The amount of ^{90}Sr in the adult rib is shown in Table 4 and Fig 2. The average of the whole series was approximated to 0.7 strontium unit (SU) (micromicrocurie of ^{90}Sr per gram of calcium). Expressed yearly, a plateau of approximately 0.6 SU is seen up until 1965, followed by a slight tendency to increase in recent years. This probably reflects the influence of repeated nuclear tests. According to Miyake and Katsuragi,[5] the fallout of ^{90}Sr and ^{137}Cs in Japan suddenly increased in 1963. In the following two years, the amount of ^{90}Sr exceeded 70% of the total fallout in the past. At the end of 1967, the presumptive cumulative fallout of ^{90}Sr in Akita district (northern Japan) was 139 millicurie/sq km, far exceeding the secondary alarm value of 90 millicurie/sq km recommended by the Atomic Energy Section of the Bureau of Science and Technology, Japan.

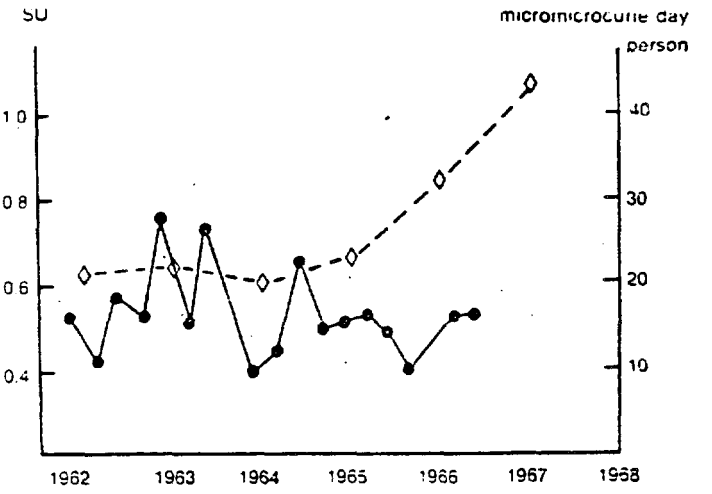
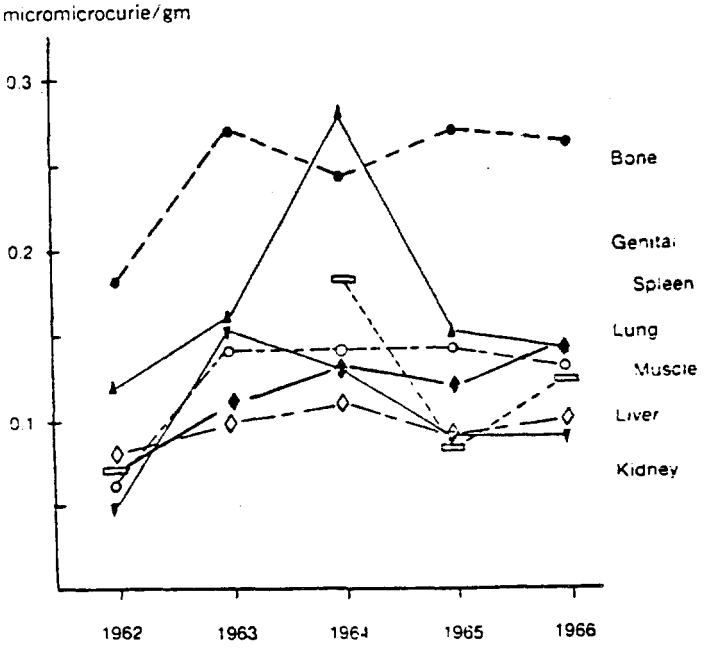


Fig 2.—Relationship of ^{90}Sr in human bone to intake of total diet. Diamonds, ^{90}Sr in human bone; circles, ^{90}Sr in standard diet.

Fig 3.—Cesium 137 of various kinds of human organs, 1962 to 1966.



The amount of ^{90}Sr varied according to the age of the subjects: those in their 20's showed the highest value of 1.3 SU, followed by 0.9 SU for those in their 30's, and 0.8 SU for the subjects in their 40's. The estimated amount in children between newborn to 9 years of age was as much as three times the average amount for adults over the age of 20.

It is evident from Table 5 and Fig 3 that ^{137}Cs was highest in the rib, 0.2 micromicrocurie/gm, with respect to the average value from 1962 to 1965. The gonads, lungs, and kidneys showed relatively low activity. The lowest concentration encountered in the kidney was 0.1 micromicrocurie/gm. The kidney and spleen showed higher values from

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1964 to 1965 than from 1962 to 1963. Although only a slight increase is noted in these organs, the gonads revealed a marked increase.

Many reports indicate an affinity of ^{137}Cs for muscular tissue. In our survey, however, this was not evident. The significant increase of ^{137}Cs in bone and genital organs would thus warrant attention to the possible dangers and, hence, to the necessity for determining the results of these hazards.

It is evident from the facts described previously that the effects of radiation on organisms has become a problem to the entire human race. Corresponding to the environmental pollution, the amount of ^{239}Pu, ^{90}Sr, and ^{137}Cs in humans was clearly observed to be greater in northern Japan, while radioactive pollution of fish, shellfish, and seaweed was similarily greater on the Japan seaside. The observed regional variation in fallout shows that there is a general tendency for the fallout to be greater in the Japan Sea than in the region bordering the Pacific Ocean.[9] The regional distribution of the fallout in northern Japan was considerably greater in winter and less in the summer, directly reflecting nuclear testing dates.

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