

PAPER X

Studies on the Metabolism of Fission Products

II. Studies on the Metabolism of the Radioisotopes Contained in the Radioactive Ashes Obtained from the No. 5 Fukuryu Maru

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INTRODUCTION

The metabolism of the fission products contained in the radioactive ashes obtained from the No. 5 Fukuryu Maru has been studied in adult mice. The radioelements studied were yttrium, cerium, praseodymium, calcium, strontium, ruthenium, rhodium, zirconium, niobium, and iodine.

MATERIALS AND METHODS

As experimental animals adult mice ranging in body weight from 10 to 16 grams were used. Usually 4 to 5 mice were used in each experiment. However, when the material was very small in amount, 2 to 3 mice were employed in one experiment. For the study of the metabolism of I^{131} , adult rats weighing about 150 grams were also used. The radioisotopes, Y^{91} , $Ce^{141,144}$, Pr^{144} , $Ru^{103,106}$, Rh^{106} , Zr^{95} and Nb^{95} were separated from the radioactive ashes using ion exchange resin Amberlite IR-120. The method of separation has been reported in detail¹⁾. The radioisotopes were diluted with physiologic saline solution so as to make the radioactivity 15,000 to 50,000 counts per minute per cc., and the pH of the solution was adjusted to 6.0. $Ru^{103,106}$ and Rh^{106} were obtained as hydrochloric acid solution by the ion exchange resin method, while Zr^{95} and Nb^{95} were obtained as the oxalate. In order to destroy the oxalate, the fraction containing Zr^{95} and Nb^{95} was treated with nitric acid, heated to dryness and dissolved in physiologic saline solution. For the separation of Y^{91} and $Ce^{141,144}$ Pr^{144} , 2mg. of inert rare earths were added as carrier to the radioactive ashes, and after the removal of silicon with hydrogen fluoride and perchloric acid, the ion exchange resin method was applied. As the fraction thus obtained contained citrate, the solution was wet ashed with perchloric acid and hydrogen peroxide in order to destroy the citrate, and Y^{91} and $Ce^{141,144}$ Pr^{144} were precipitated from neutral medium. The precipitate was dissolved in hydrochloric acid, diluted with physiologic

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saline solution, and the pH was adjusted to 6.0. For the study of the metabolism of radiostrontium, radiocalcium and radioiodine, the radioisotopes supplied by the U. S. AEC were used. The radioisotopes were given by mouth or subcutaneously to adult mice. For oral administration, a stomach tube, 1 mm. in diameter, attached to a syringe was used in order to introduce the material directly into the stomach. In case of subcutaneous injection, the material was injected between the two shoulder blades in the back of the animals. For the disinfection of the injection site tincture of iodine was used in order to prevent the animals from licking the injection site. The animals were sacrificed 12 hours following the oral administration or 4 and 12 hours following the subcutaneous injection, (in case of Sr^{90} 12 and 24 hours and 4, 14 and 30 days following the injection) and the liver, lungs, kidneys, spleen, blood, digestive tract (including the stomach, small and large intestines together with their contents) and bones (both femurs, tibias and fibulas including the bone marrow) were removed, weighed, and wet ashed with perchloric acid and hydrogen peroxide. The samples were transferred into glass dishes, 3 cm. in diameter, dried,

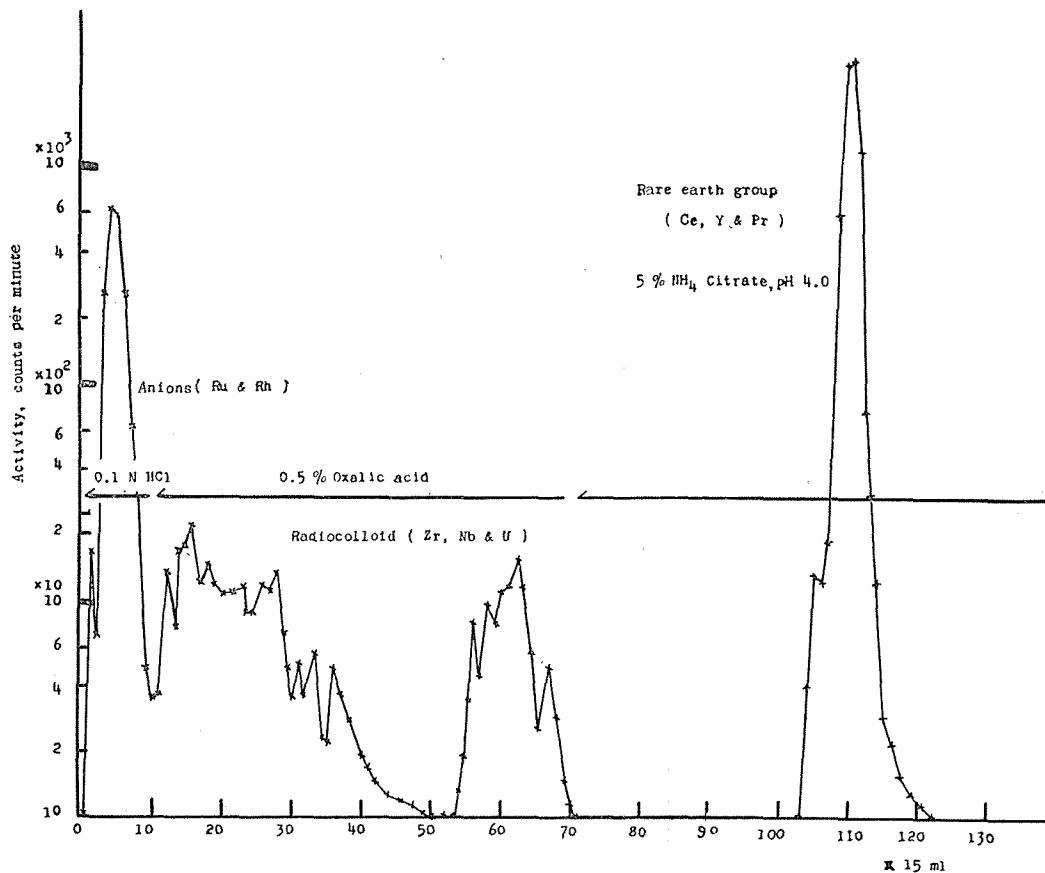


Fig. 1. Elution curve of cation exchange group separation for animal experiments, using Amberlite IR-120 (60 mesh, 1 cm. ϕ \times 70 cm.).

and the radioactivity was measured with a Geiger-Mueller counter and a "100" scaler. As a standard, the same amount of the material as the administered dose (or 1/100 of the administered dose) was treated in the same way as the samples and the radioactivity was measured under the same conditions. For the measurement of radioiodine a scintillation counter was used.

The results were expressed as per cent of the administered dose per gram tissue and per whole organ.

In case of Ru^{103,106} Rh¹⁰⁶ the tissues were spread uniformly between two sheets of thin paper and dried at 37°C. in an incubator to prevent ruthenium and rhodium

Fig. 2. Al absorption curve for Y⁹¹.

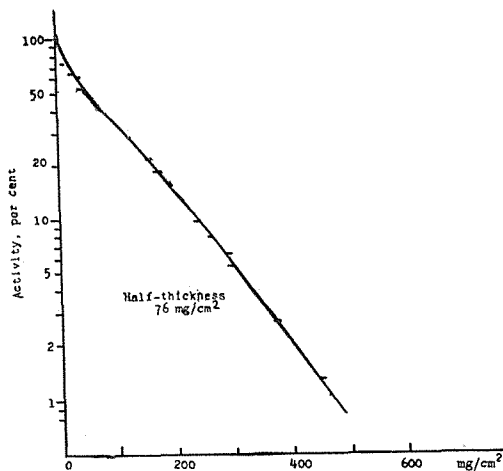


Fig. 3. Al absorption curve for Ce^{141,144} and Pr¹⁴⁴.

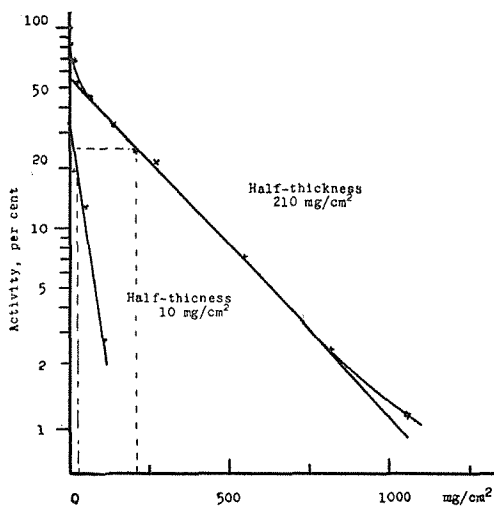
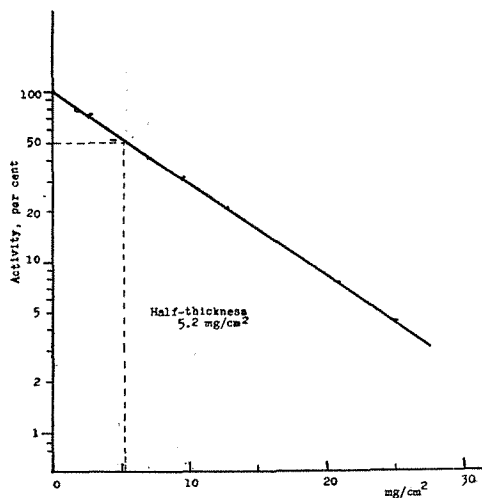


Fig. 4. Al absorption curve for Ca⁴⁵.



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from being lost by evaporation during wet ashing with perchloric acid. For the radioactivity measurement of the radioisotopes with low energy beta particles, such as Ca^{45} , $\text{Ru}^{103,106}$, Rh^{106} and Zr^{95} , Nb^{95} , self absorption correction was made by measuring the weight of the samples.

As the radiostrontium used in our experiments was a mixture of Sr^{89} and Sr^{90} , Y^{90} was contained as a daughter product of Sr^{90} . Although Y^{90} might have exerted some influence upon the metabolism of radiostrontium, radioactivity measurement was made only once 24 hours after the preparation of the samples, and no further analysis was made. Fig. 1 shows the elution curve obtained by the ion exchange resin method, and Figs. 2~8 show the aluminium absorption curves²⁾ of the radio-

Fig. 5. Al absorption curve for $\text{Sr}^{89,90}$.

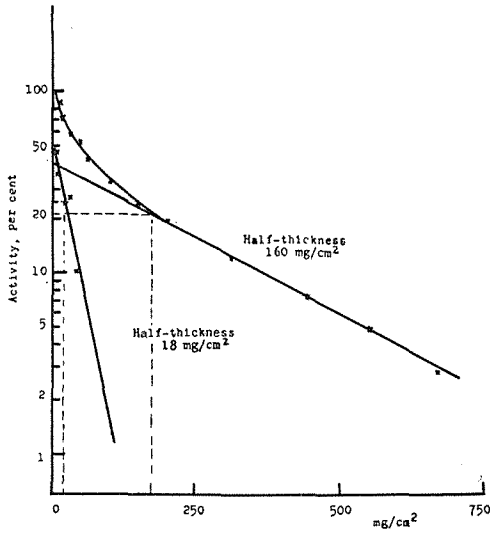


Fig. 6. Al absorption curve for $\text{Ru}^{103,106}$ and Rh^{106} .

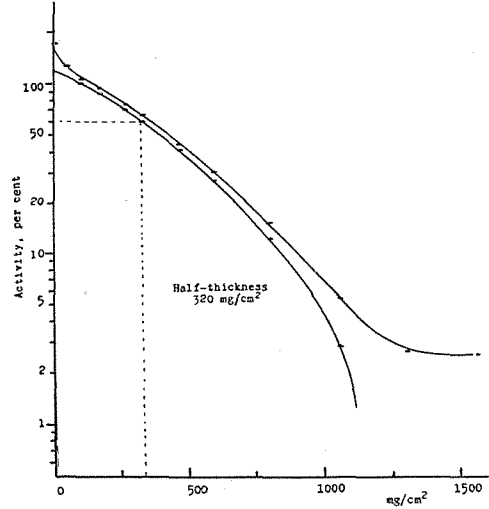
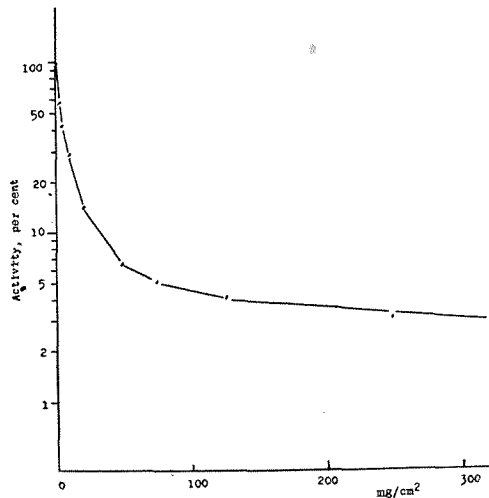
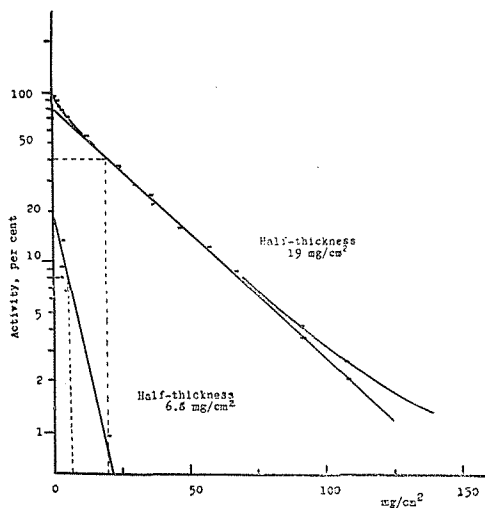


Fig. 7. Al absorption curve for Zr^{95} and Nb^{95} .



isotopes used in our experiments.

Fig. 8. Al absorption curve for I^{131} .



(1) Y^{91} (Table 1 and Figs. 9, 10, 11)

The absorption of Y^{91} from the digestive tract following the oral administration was poor. When administered subcutaneously, Y^{91} was deposited chiefly in the bones. Four hours following the subcutaneous injection, the radioactivity per gram tissue of the bones was the highest, followed by that of the digestive tract, kidneys and liver in this order. The excretion of Y^{91} occurred through the kidneys and digestive tract.

Fig. 9. Distribution of Y^{91} in the tissues of the mouse 12 hours following oral administration (per gram).

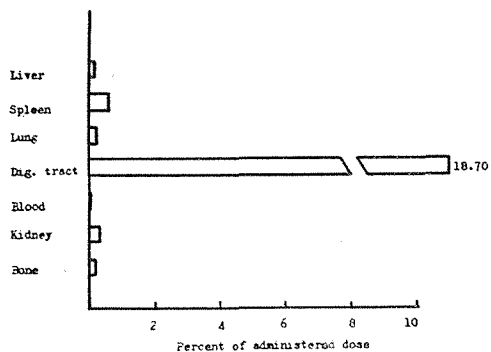
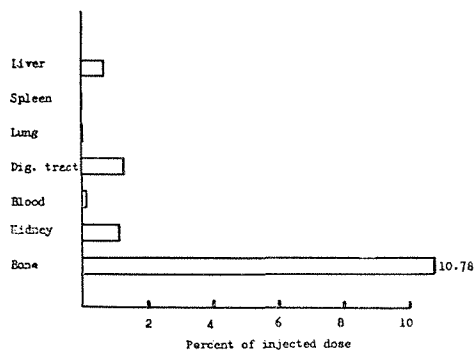


Fig. 10. Distribution of Y^{91} in the tissues of the mouse 4 hours following subcutaneous injection (per gram).



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Table 1. Distribution of Y⁹¹ and Ce^{141,144} Pr¹⁴⁴ in the tissues of the mouse

Element		Y ⁹¹	Y ⁹¹		Ce ^{141,144} Pr ¹⁴⁴
Method of administration		Oral admin.	Subcutan. injection		Subcutan. injection
Time after administration		12 hrs.	4 hrs.	12 hrs.	4 hrs.
Number of animals		5	5	3	2
Liver	p. w. o.	0.19±0.07	0.37±0.09	0.49±0.09	0.08±0.02
	p. g. t.	0.21±0.06	0.67±0.39	0.75±0.14	0.13±0.02
Spleen	p. w. o.	0.04±0.01	0.00±0.00	0.01±0.01	0.01±0.005
	p. g. t.	0.62±0.18	0.00±0.00	0.20±0.19	0.13±0.12
Lung	p. w. o.	0.04±0.02	0.01±0.01	0.05±0.03	0.01±0.01
	p. g. t.	0.24±0.12	0.07±0.04	0.37±0.23	0.11±0.11
Digest. tract	p. w. o.	53.87±11.84	2.52±1.83	2.88±1.02	0.25±0.14
	p. g. t.	18.70±3.73	1.31±0.90	1.26±0.50	0.11±0.04
Blood	p. g.	0.05±0.01	0.18±0.14	0.04±0.01	0.12±0.025
Kidney	p. w. o.	0.06±0.03	0.16±0.09	0.06±0.03	0.02±0.015
	p. g. t.	0.27±0.11	1.15±0.73	0.43±0.19	0.075±0.075
Bone	p. w. s.	0.02±0.01	2.98±1.26	0.47±0.12	0.11±0.015
	p. g. t.	0.15±0.07	10.78±4.70	2.01±0.59	0.42±0.035

p. w. o. per whole organ
 p. g. t. per gram tissue
 p. g. per gram
 p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

Fig. 11. Distribution of Y⁹¹ in the tissues of the mouse 12 hours following subcutaneous injection (per gram).

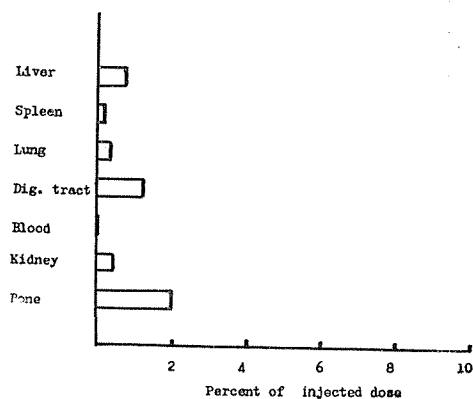
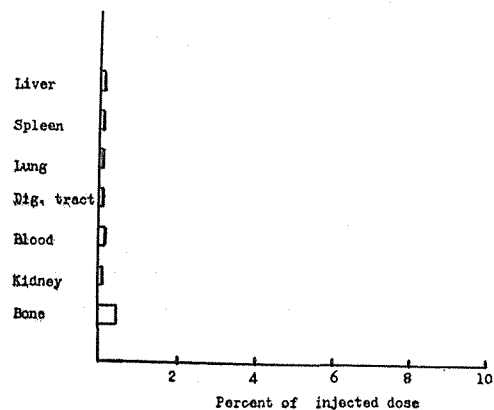


Fig. 12. Distribution of Ce^{141,144} and Pr¹⁴⁴ in the tissues of the mouse 4 hours following subcutaneous injection (per gram).



(2) $Ce^{141,144}$, Pr^{144} (Table 1 and Fig. 12)

Four hours following the subcutaneous injection of $Ce^{141,144}$ and Pr^{144} the radioactivity per gram tissue was highest in the bones. However, the deposition of $Ce^{141,144}$ and Pr^{144} in the bones was much less as compared with that of Y^{91} . Small amounts of radioactivity were also found in the liver, spleen, digestive tract and kidneys.

(3) Ca^{45} (Table 2 and Figs. 13, 14)

The absorption of Ca^{45} from the digestive tract was moderately good, and less than 8 % of the administered dose remained in the gastrointestinal tract 12 hours following the oral administration. The main deposition site of Ca^{45} was the bones following both the oral and subcutaneous administrations. The excretion of Ca^{45} occurred through the kidneys and the digestive tract.

Table 2. Distribution of Ca^{45} in the tissues of the mouse

Element		Ca^{45}	Ca^{45}
Method of administration		Oral admin.	Subcutan. injection
Time after administration		12 hrs.	4 hrs.
Number of animals		4	5
Liver	p. w. o.	0.73±0.27	1.37±0.26
	p. g. t.	1.18±0.26	1.99±0.44
Spleen	p. w. o.	0.11±0.02	0.21±0.05
	p. g. t.	0.85±0.05	3.46±0.81
Lung	p. w. o.	0.25±0.16	0.07±0.005
	p. g. t.	1.44±0.84	0.36±0.03
Digest. tract	p. w. o.	6.81±0.19	6.99±0.68
	p. g. t.	3.76±0.90	2.96±0.28
Blood	p. g.	2.43±0.33	4.15±0.94
Kidney	p. w. o.	0.08±0.04	1.36±0.42
	p. g. t.	0.43±0.23	6.13±2.04
Bone	p. w. s.	6.29±0.63	7.71±1.19
	p. g. t.	24.90±3.5	29.25±4.96

p. w. o. per whole organ
 p. g. t. per gram tissue
 p. g. per gram
 p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

(4) $Sr^{90,90}$ (Table 3 and Figs. 15, 16, 17, 18, 19)

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Fig. 13. Distribution of Ca⁴⁵ in the tissues of the mouse 12 hours following oral administration (per gram).

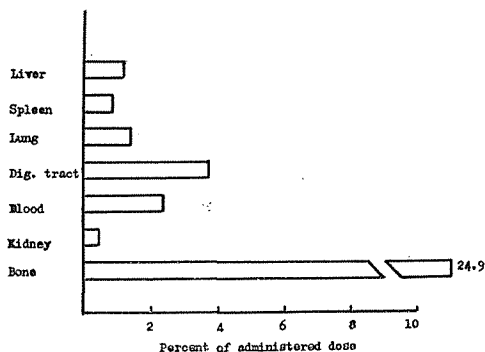
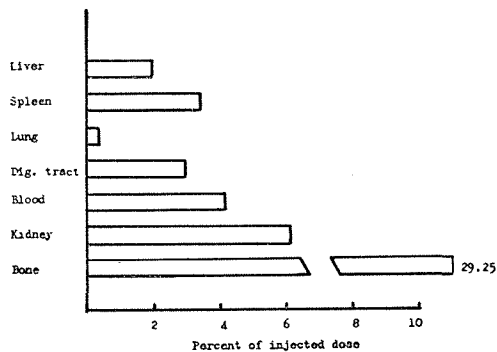


Fig. 14. Distribution of Ca⁴⁵ in the tissues of the mouse 4 hours following subcutaneous injection (per gram).



Twelve hours following the oral administration, Sr^{90,90} was deposited chiefly in the bones. The absorption of Sr^{90,90} from the digestive tract was much better than that of Y⁹¹ and Ce^{141,144}, Pr¹⁴⁴. Four hours following the subcutaneous injection, the deposi-

Table 3. Distribution of Sr^{90,90} in the tissues of the mouse

Method of administration		Oral admin.	Subcutan. injection	Subcutan. injection	Subcutan. injection	Subcutan. injection	Subcutan. injection	Subcutan. injection
Time after administration		12 hrs.	4 hrs.	12 hrs.	24 hrs.	4 days	14 days	30 days
Number of animals		5	5	5	5	5	5	5
Liver	p. w. o.	0.06±0.01	0.83±0.11	0.80±0.06	0.67±0.09	0.29±0.02	0.04±0.007	0.013±0.004
	p. g. t.	0.08±0.02	1.38±0.13	1.28±0.10	1.43±0.14	0.57±0.04	0.06±0.013	0.018±0.002
Spleen	p. w. o.	0.007±0.003	0.02±0.003	0.02±0.007	0.01±0.00	0.01±0.002	0.00±0.00	0.00±0.00
	p. g. t.	0.14±0.07	0.35±0.05	0.28±0.07	0.20±0.02	0.13±0.023	0.00±0.00	0.00±0.00
Lung	p. w. o.	0.02±0.009	0.12±0.02	0.08±0.006	0.04±0.01	0.02±0.006	0.002±0.002	0.001±0.001
	p. g. t.	0.16±0.05	1.12±0.24	0.73±0.05	0.27±0.04	0.18±0.031	0.02±0.006	0.004±0.002
Digest. tract	p. w. o.	10.51±5.05	2.93±0.39	3.57±0.55	1.54±0.12	0.37±0.10	0.08±0.012	0.027±0.017
	p. g. t.	4.15±2.06	1.05±0.18	1.73±0.06	0.85±0.17	0.18±0.04	0.05±0.007	0.01±0.007
Blood	p. g.	0.14±0.03	0.90±0.29	0.59±0.12	0.38±0.09	0.08±0.03	0.02±0.005	0.001±0.0002
Kidney	p. w. o.	0.02±0.007	0.32±0.026	0.31±0.02	0.20±0.04	0.11±0.006	0.008±0.002	0.001±0.0001
	p. g. t.	0.16±0.06	0.72±0.05	2.51±0.28	1.40±0.41	0.97±0.037	0.02±0.009	0.005±0.002
Bone	p. w. s.	1.05±0.20	2.67±0.32	3.69±0.46	3.12±0.44	2.69±0.15	1.81±0.27	1.24±0.17
	p. g. t.	5.90±0.96	19.64±4.12	18.14±2.50	18.39±1.13	14.24±0.79	10.33±0.67	7.52±0.84

p. w. o. per whole organ
 p. g. t. per gram tissue
 p. g. per gram
 p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

Fig. 15. Distribution of $Sr^{89,90}$ in the tissues of the mouse 12 hours following oral administration (per gram).

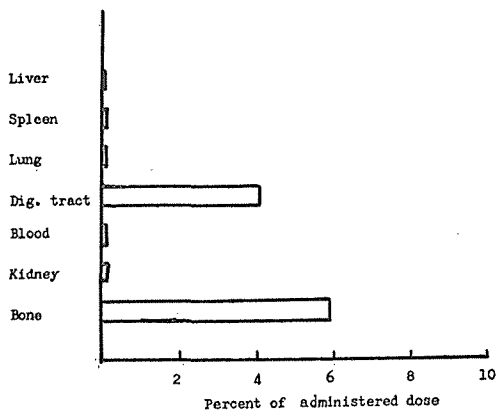


Fig. 16. Distribution of $Sr^{89,90}$ in the tissues of the mouse 4 hours following subcutaneous injection (per gram).

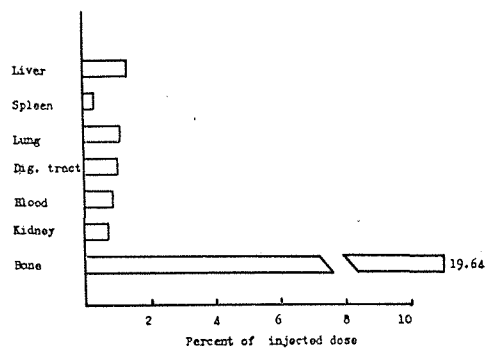


Fig. 17. Distribution of $Sr^{89,90}$ in the tissues of the mouse 12 hours following subcutaneous injection (per gram).

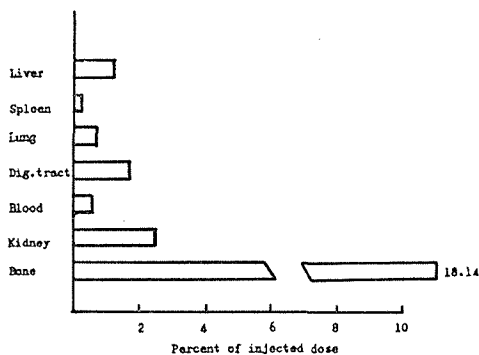
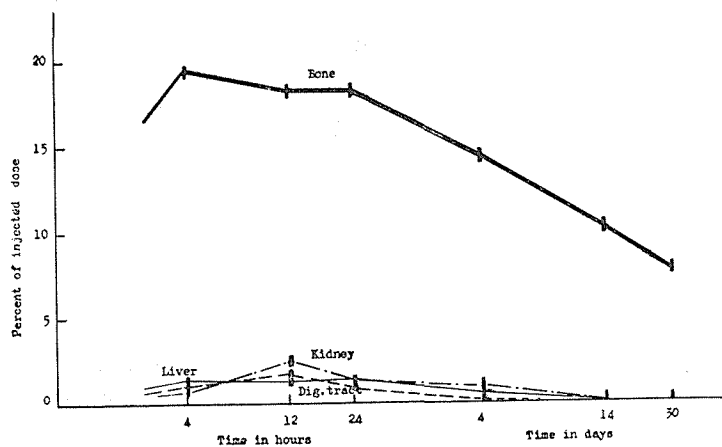


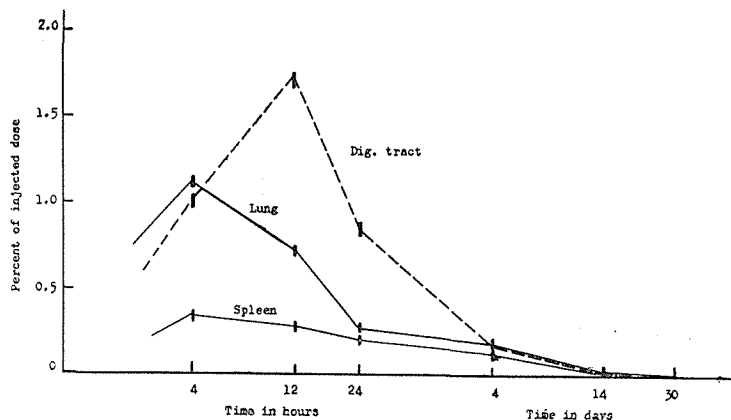
Fig. 18. Distribution of $Sr^{89,90}$ in the tissues of the mouse 4, 12 and 24 hours and 4, 14 and 30 days following subcutaneous injection (per gram).



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tion of $\text{Sr}^{89,90}$ was also highest in the bones. The excretion of $\text{Sr}^{89,90}$ occurred through the kidneys and digestive tract. The radiostrontium, deposited in the tissues other than the bones, was eliminated relatively rapidly, while the elimination of the radiostrontium deposited in the bones was very slow, and even 30 days following the subcutaneous injection a considerable amount of the radiostrontium was still remaining in the bones.

Fig. 19. Distribution of $\text{Sr}^{89,90}$ in the tissues of the mouse 4, 12 and 24 hours and 4, 14 and 30 days following subcutaneous injection (per gram).



(5) $\text{Ru}^{103,106}$, Rh^{106} (Table 4 and Figs. 20, 21)

The absorption of $\text{Ru}^{103,106}$ and Rh^{106} from the digestive tract was poor. The deposition of $\text{Ru}^{103,106}$ and Rh^{106} in the bones was also poor. $\text{Ru}^{103,106}$ and Rh^{106} showed a tendency to be deposited in the kidneys, liver, lungs and spleen.

(6) Zr^{95} , Nb^{95} (Table 4 and Figs. 22, 23)

The absorption of Zr^{95} and Nb^{95} from the digestive tract was not good. Four hours following the subcutaneous injection 11.51% of the administered dose was excreted mainly through the kidneys, and partly through the digestive tract, and only a small amount of the administered dose was deposited in the bones. Zr^{95} and Nb^{95} showed a tendency to be deposited in the spleen and lungs. Zr^{95} and Nb^{95} tended to form colloidal aggregates, and the uptake of these radioisotopes by the spleen and lungs might be partly explained by the phagocytosis of the elements by the reticuloendothelial cells in these organs. When silicon is present with Zr^{95} and Nb^{95} , this tendency would be enhanced, because the colloid aggregates of Zr^{95} and Nb^{95} are absorbed on the silicon.

(7) I^{131} (Table 5 and Figs. 24, 25)

The absorption of I^{131} from the digestive tract was excellent. When injected subcutaneously, the excretion occurred through the kidneys and partly through the

Table 4. Distribution of Ru^{103,106}, Rh¹⁰⁶ and Zr⁹⁵, Nb⁹⁵ in the tissues of the mouse

Element		Ru ^{103,106} , Rh ¹⁰⁶		Zr ⁹⁵ , Nb ⁹⁵	
		Oral admin.	Subcutan. injection	Oral admin.	Subcutan. injection
Method of administration					
Time after administration		12 hrs.	4 hrs.	12 hrs.	4 hrs.
Number of animals		2	2	2	2
Liver	p. w. o.	0.16	0.22±0.06	0.02±0.00	0.08±0.01
	p. g. t.	0.25	0.49±0.11	0.03±0.00	0.11±0.04
Spleen	p. w. o.	0.05	0.04±0.04	0.00±0.000	0.05±0.01
	p. g. t.	0.53	0.27±0.27	0.00±0.00	0.59±0.02
Lung	p. w. o.	0.11	0.06±0.02	0.03±0.01	0.03±0.01
	p. g. t.	0.72	0.34±0.06	0.25±0.03	0.19±0.09
Digest. tract	p. w. o.	28.83	0.58±0.14	36.49±4.81	0.17±0.04
	p. g. t.	8.24	0.28±0.12	19.70±2.50	0.09±0.03
Blood	p. g.	0.36	0.37±0.24	0.05±0.02	0.13±0.00
Kidney	p. w. o.	0.11	0.13±0.08	0.02±0.01	0.04±0.00
	p. g. t.	0.56	0.52±0.29	0.15±0.10	0.27±0.00
Bone	p. w. s.	0.28	0.24±0.15	0.04±0.02	0.20±0.04
	p. g. t.	0.62	0.78±0.51	0.25±0.05	1.00±0.20
Excreta	total	48.36	2.70±0.15		11.51

p. w. o. per whole organ
 p. g. t. per gram tissue
 p. g. per gram
 p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose ± the standard error of the mean.

Fig. 20. Distribution of Ru^{103,106} and Rh¹⁰⁶ in the tissues of the mouse 12 hours following oral administration (per gram).

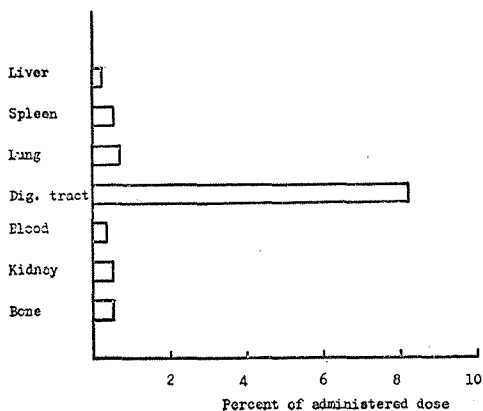
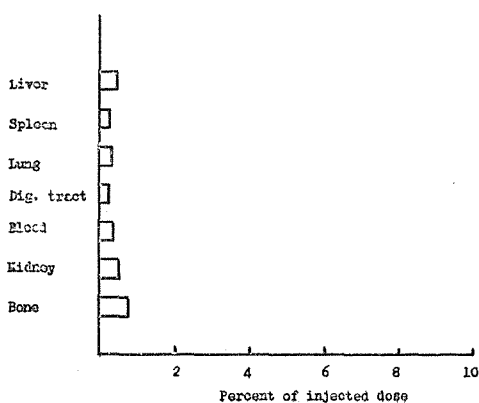


Fig. 21. Distribution of Ru^{103,106} and Rh¹⁰⁶ in the tissues of the mouse 4 hours following subcutaneous injection (per gram).



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Fig. 22. Distribution of Zr^{95} and Nb^{95} in the tissues of the mouse 12 hours following oral administration (per gram).

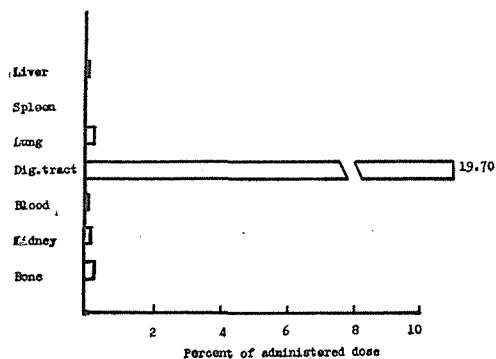


Fig. 23. Distribution of Zr^{95} and Nb^{95} in the tissues of the mouse 4 hours following subcutaneous injection (per gram).

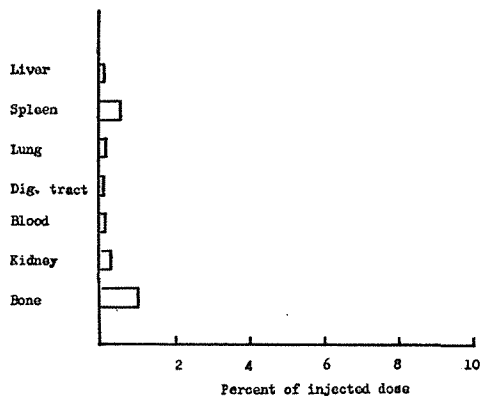


Table 5. Distribution of I^{131} in the tissues of the mouse and rat

Element		I^{131}	I^{131}
Method of administration		Oral admin.	Subcutan. injection
Time after administration		12 hrs.	4 hrs.
Number of animals		2 mice	3 rats
Liver	p. w. o.	0.29 ± 0.02	2.81 ± 1.87
	p. g. t.	0.30 ± 0.03	0.46 ± 0.29
Spleen	p. w. o.	0.02 ± 0.00	0.18 ± 0.05
	p. g. t.	0.48 ± 0.02	0.36 ± 0.08
Lung	p. w. o.	0.08 ± 0.01	1.86 ± 1.06
	p. g. t.	0.50 ± 0.01	1.77 ± 0.88
Digest. tract	p. w. o.	2.47 ± 0.71	36.45 ± 0.63
	p. g. t.	1.10 ± 0.42	2.98 ± 0.04
Blood	p. g.	1.37 ± 0.56	1.02 ± 0.34
Kidney	p. w. o.	0.20 ± 0.03	0.96 ± 0.32
	p. g. t.	1.52 ± 0.38	0.71 ± 0.25
Bone	p. w. s.	0.17 ± 0.01	1.78 ± 0.04
	p. g. t.	1.06 ± 0.13	0.60 ± 0.02
Thyroid	p. g. t.		380

p. w. o. per whole organ

p. g. t. per gram tissue

p. g. per gram

p. w. s. per whole sample (both femurs, tibias and fibulas)

Values are presented as group means of the percent of the administered dose \pm the standard error of the mean.

digestive tract. Four hours following the subcutaneous injection of I^{131} in adult rats the deposition of I^{131} in the thyroid amounted to 380% of the administered dose per gram tissue.

Fig. 24. Distribution of I^{131} in the tissues of the mouse 12 hours following oral administration (per gram).

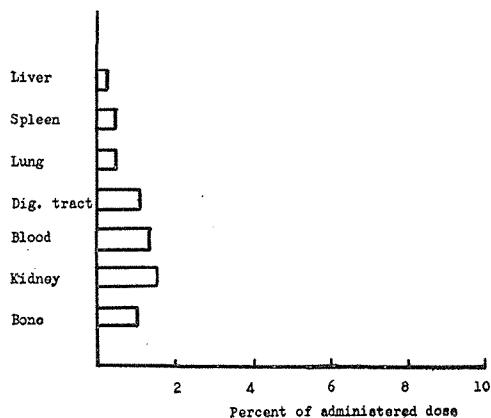
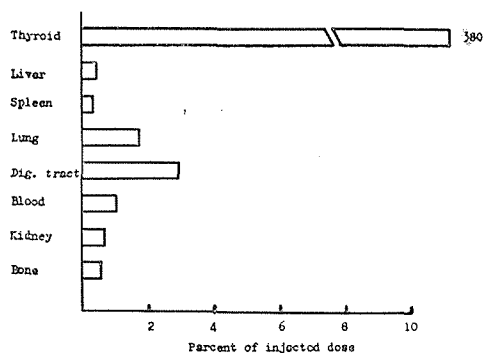


Fig. 25. Distribution of I^{131} in the tissues of the rat 4 hours following subcutaneous injection (per gram).



DISCUSSION

The metabolism of fission products has been studied by many investigators^{3,4,5,6,7} using the radioisotopes produced in the cyclotron or uranium pile reactor. In our experiments most of the fission products studied were separated from the radioactive ashes collected from the No. 5 Fukuryu Maru. For the separation of rare earths inert elements were added as carrier. However, the amount of the carrier added was so small that the influence of the carrier upon the metabolism of the radioelements would have been negligible. For the separation of the radioisotopes other than the rare earths no carrier was added. As the material used in our experiments was originally separated from the dust collected from the deck of the No. 5 Fukuryu Maru, there was a possibility that the material contained many impurities such as iron, copper, zinc, calcium, silicon, chromium, etc. Although these impurities were removed as much as possible, some of them might have remained in the test dose and exerted some influence upon the metabolism of the fission products. Be that as it may, the results obtained in our experiments were similar to those reported by other investigators. Among the radioisotopes studied, radioiodine, radio-calcium and radiostrontium were readily absorbed from the digestive tract, while the absorption of the rare earths from the digestive tract was poor. The radioelements which accumulated chiefly in the bones were radiocalcium, radiostrontium and radioyttrium. Radiostrontium rapidly concentrates in the skeleton, and it is very slowly excreted, with a biological half-life of 3.9×10^3 days.¹⁰ Therefore,

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among the fission products studied, radiostrontium appears to be the most important radioisotope, which accumulates in the skeleton and causes biological hazards. The heavy metals such as radioruthenium and radiorhodium tended to be distributed in the kidneys, liver, lungs and spleen.

SUMMARY

1) The metabolism of the radioisotopes found in the radioactive ashes collected from the No. 5 Fukuryu Maru has been studied in adult mice. The radioisotopes studied were Y^{91} , $Ce^{141,144}$, Pr^{144} , Ca^{45} , $Sr^{89,90}$, $Ru^{103,106}$, Rh^{106} , Zr^{95} , Nb^{95} and I^{131} .

2) Among the radioisotopes studied, radiostrontium, radiocalcium and radioyttrium accumulated chiefly in the bones, and the elimination of radiostrontium from the bones was very slow.

3) When administered by mouth, radiostrontium and radiocalcium were readily absorbed from the digestive tract, while the absorption of radioyttrium from the digestive tract was poor.

4) Among the radioisotopes studied, radiostrontium appears to be the most important radioisotope as a cause of biological hazards, when the radioactive ashes are taken by mouth.

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