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Fallout Radioactivity in Excreta and Diet

——The Levels at Tokai, Japan in 1962 to 1963——

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Japan Atomic Energy Research Institute

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ABSTRACT

The fallout levels of fission products such as ^{90}Sr, ^{137}Cs, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru, in excreta and diet were measured. In connection with ^{137}Cs and ^{90}Sr, the amounts of stable potassium and calcium in the same specimens were also analysed. The volume and weight of the samples were examined to get the mean value and the range.

The experiment started in May 1962 and ended in February 1963. During the period, seven samplings were made. In each sampling, excreta were collected over five consecuitive days from five normal subjects, who, except one, received the same diet. An exact duplicate of the diet was analysed at the same time.

A large NaI(Tl) crystal scintillator and a 100 channel pulse-height analyser were used to determine the radioactivities of γ-emitters in ashed samples, while ^{90}Sr and ^{137}Cs were analysed radiochemically.

In general, the content of ^{95}Nb in the diet was highest among all the radionuclides, reaching about 60 $\mu\mu$c/day/person in February 1963. In urine samples, detectable amounts of ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru were not found, in contrast with the considerable amounts in feces. The level of ^{90}Sr in the diet varied between 7.0 and 13 $\mu\mu$c/day/person during the period, and that of ^{137}Cs, between 13 and 35 $\mu\mu$c/day/person. However, in 24-hr urine samples, about 1 $\mu\mu$c of ^{90}Sr was always found irrespective of the contents in the diet. The activities of ^{137}Cs in the same urine specimens, however, changed between 10 and 31 $\mu\mu$c corresponding to the contents in the diet.

The levels of ^{90}Sr, ^{137}Cs and $^{95}Zr + ^{95}Nb$ involved in the diet varied during the period according to the changes of the dry fallout levels in the atmosphere.

Moreover, the average daily volume of urine for individuals was 950 to 1300 ml.

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排泄物中および食品中におけるフォールアウト放射能

―― 1962年〜1963年の東海における放射能強度測定結果 ――

要　旨

^{90}Sr，^{137}Cs，^{95}Zr，^{95}Nb，^{103}Ru および^{106}Ruのような，核分裂生成物の排泄物中および食品中におけるフォールアウト放射能のレベルを測定した。安定KとCaは，それぞれ^{137}Cs と^{90}Srと関連があるので，同じ標本について定量した。標本の容量と重量も調べ，平均値と範囲を求めた。

本実験は1962年の5月に開始し，1963年の2月に終了した。この期間中，7回にわたってサンプリングをおこなった。排泄物は各サンプリングごとに連続する5日間，5人の正常な人間から採取した。サンプリング期間中は5人のうち1人を除いて皆同じ食事をとり，それと同じ食事を別にサンプリングして分析をおこなった。

γ線を出す核種の定量には，灰化した試料を用い，NaI（Tl）の大きな結晶と100チャンネルの波高分析器を使用した。一方，^{90}Srと^{137}Cs の定量は放射化学分析によった。

一般に，食品中の^{95}Nbの量がすべての核種中最高であって，1963年の2月には約60 μμc／日／人に達した。

尿中には^{95}Zr，^{95}Nb，^{103}Ruおよび^{106}Ru は存在せず，糞中に，かなりの量が見出されたのと対照的であった。食品中の^{90}Srのレベルはこの期間中，7.0から13μμc/日／人 の間変化し，^{137}Cs は13から35 μμc／日／人 まで変動した。しかし，24時間尿中の^{90}Sr の量は常に約1μμcであって，食品中の量と直接の関係はなかった。ところが尿中の^{137}Cs の量は10から31 μμc／日／人 までの間を変化し，食品中の値に対応していた。

食品中に含まれる^{90}Sr，^{137}Csおよび^{95}Zr＋^{95}Nb のレベルは，この期間中，大気中のドライ・フォールアウトのレベルの変化に呼応して変動した。

また，個人別に平均した尿の1日あたりの排泄量は 950ないし1,300 mℓ であった。

1963年12月

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1. INTRODUCTION

The assessment of internal exposure is usually performed on excretion analysis. Fallout radioactivities in excreta, which work as the background, vary with individuals and with time, according to differences in dietary intake. This variation becomes a source of error in determining occupational exposures from excretion analysis. Accordingly, for the accurate evaluation of the dose, it is necessary to determine the mean value and range of the fallout radioactivities.

Moreover, the mean value and range of daily excretion in terms of volume and weight also have to be known beforehand, since in the routine sampling of excreta for bioassay, it is difficult to collect 24-hr samples and the radioactivities in them must be estimated from spot samples.

In this study, fallout radioactivities, volume, and weight of excreta were determined, using specimens collected from five normal male subjects for seven periods of five days each. Furthermore, to elucidate the correlation between the radioactivities in excreta and those in diet, the subjects, except one, received the same diet during each period and the radioactivities in the diet were analysed.

In the present paper, the levels of fission products such as ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru and ^{137}Cs, and natural ^{40}K, in diet, urine, and feces are described. In addition, reported are the daily volume and weight of excreta and diet as well as the amounts of stable potassium and calcium in them.

2. METHODS

In order to save time, the physical and chemical procedures for treatment of samples were simplified as much as possible.

Outlines of the procedures are as follows. The *diet* and *feces* were dry ashed, and the radioactivities of ^{40}K, ^{95}Zr, ^{95}Nb, ^{103}Ru or ^{106}Ru were measured directly by γ-spectrometry, while ^{90}Sr and ^{137}Cs were determined radiochemically from the ashes. For *urine* samples, a co-precipitation method was used to avoid laborious wet-ashing procedures. Most fission radionuclides in urine were co-precipitated on the basic phosphates of calcium and magnesium(1)(2). The γ-emitters in the phosphate precipitate were determined by γ-spectrometry. However, ^{90}Sr and ^{137}Cs were analysed radiochemically from the phosphate precipitate and the filtrate, respectively.

As the radioactivities in the daily samples were, in general, too weak to determine accurately, five-day samples were collected and measured as a whole to get more accurate results.

2.1 The Subjects and Sampling Periods

Table 1 shows the age, height and weight of the five volunteers.

Subject I lived in Mito city, Ibaraki Pref., and Subjects II ~ V, in a JAERI dormitory at Tokai village near Mito. The Subject I took his breakfast and supper at his home, and lunch, at the JAERI cafeteria. The other subjects took their breakfast and supper at the JAERI dormitory, and lunch, at the same cafeteria. Besides the ordinary meals, all subjects took two bottles of milk per day (one is the usual white milk and the other is a fruit-milk, each 180 ml). Except for the first and the last periods, fruits and cakes were not taken by the subjects.

Collection of the diet and excreta began in May 1962 and ended in February 1963. The details of the sampling periods are shown in the upper part of Table 3 (see page 28).

2.2 Sampling Procedure and Treatment for γ-spectrometry

Diet An exact duplicate of the diet was collected in a polyethylene sack. After weighing, the sample was air dried at 110 ~ 130°C overnight. The dried sample was weighed, and ashed in a muffle furnace at 550 ~ 600°C for about 10 hr. The weight of ashes was measured and transferred to γ-spectrometry (see section 2.3).

Urine Urine was collected in polyethylene bottles, and the total volume of 24-hr samples was recorded. An aliquot of the sample was removed for assay of calcium and potassium. The remainder was acidified with about 50 ml of concentrated nitric acid, and solutions of strontium and cesium carriers (each contained 10 mg) were added. The mixture was heated to 80 ~ 90°C and (1:1) ammonium hydroxide solution was added until the pH reached about 9, where calcium and magnesium were precipitated as phosphates. After standing the solution overnight, the supernatant solution was decanted, and the precipitates were centrifuged. The same precipitation procedure was repeated. The precipitates were dried at 110°C, and weighed. Subsequently, γ-spectrometry of the precipitates was carried out.

Feces Each fecal sample was wrapped in cellophane paper and then placed in a polyethylene sack. After transferring the sample on a quartz dish, a small amount of alcohol was poured onto it. The quartz dish was placed in the muffle furnace and alcohol was lighted in order to burn the cellophane paper and polyethylene sack. The sample was ashed at 550 ~ 600°C for about 10 hr, and the ashes were weighed.

Feces were not excreted every day in some volunteers because of constipation. However, in the present study, the daily weight of feces and the daily content of nuclides were obtained by dividing the total amount during five days by five.

2.3 Gamma-spectrometry

The apparatus used was a Sunvic 100 channel pulse-height analyser with a 5 × 4"NaI(Tl) crystal. To reduce the background, the detector, phototubes and samples were shielded by 20 cm of steel and 0.3 cm of lead.

The samples were put into small polyethylene sacks to get definite geometries. Gamma-spectra were obtained by counting for 30 minutes.

The amount of nuclides was calculated from the peak efficiency (ϵ_p) and the fraction of γ-disintegration at the given energy band. The peak efficiency was measured with standard sources which had the same dimensions as the samples.

Table 2 shows the sample size, the distance between sample and detector, the energy band, the peak efficiency and the detectable limits of the nuclides*.

The amounts of nuclides are expressed by the following equations.

For ^{40}K

$$^{40}K(\mu\mu c) \quad \frac{n_p(^{40}K)}{\epsilon_p(^{40}K)} \cdot \frac{1}{\gamma(^{40}K)} \cdot \frac{1}{2.22} \quad \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

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* The levels of the detectable limits vary according to the Compton scattering caused by co-existed radionuclides, fluctuation of the background activity, and the measuring time.

where

n_p : counting rate of the nuclide at the given energy band (cpm),

ε_p : peak efficiency of the nuclide at the given energy band (cpm/dpm),

γ : γ-disintegration ratio of the nuclide at the given energy band, and

2.22 : conversion factor of dpm to $\mu\mu$c.

For ^{95}Zr and ^{95}Nb Assuming that a radiochemical equilibrium has been established between ^{95}Zr (parent) and ^{95}Nb (daughter), the ratio of the numbers of atoms, ^{95}Nb/^{95}Zr, is calculated as follows:

$$\frac{^{95}\mathrm{Nb}}{^{95}\mathrm{Zr}} = \frac{\lambda(^{95}\mathrm{Zr})}{\lambda(^{95}\mathrm{Nb}) - \lambda(^{95}\mathrm{Zr})} = 1.2 \quad \dots\dots\dots\dots\dots\dots \quad (2)$$

where $\lambda(^{95}\mathrm{Zr})$ and $\lambda(^{95}\mathrm{Nb})$ are the decay constants of ^{95}Zr and ^{95}Nb, respectively.

The ratio (k) of the activities is, then, obtained using the above figure 1.2,

$$k = \frac{^{95}\mathrm{Nb}(\mu\mu c)}{^{95}\mathrm{Zr}(\mu\mu c)} = \frac{^{95}\mathrm{Nb}\ \lambda(^{95}\mathrm{Nb})}{^{95}\mathrm{Zr}\ \lambda(^{95}\mathrm{Zr})} = \frac{1.2\cdot\lambda(^{95}\mathrm{Nb})}{\lambda(^{95}\mathrm{Zr})} = 2.2 \quad \dots\dots \quad (3)$$

Accordingly, the amount of ^{95}Nb is represented as follows;

$$^{95}\mathrm{Nb}(\mu\mu c) = \frac{n_p(^{95}\mathrm{Zr} + {}^{95}\mathrm{Nb}) - f_1(^{40}\mathrm{K})n_p(^{40}\mathrm{K})}{\varepsilon_p(^{95}\mathrm{Zr} + {}^{95}\mathrm{Nb})} \times \frac{1}{\frac{\gamma(^{95}\mathrm{Zr})}{k} + \gamma(^{95}\mathrm{Nb})} \cdot \frac{D}{2.22} \quad \dots\dots\dots\dots \quad (4)$$

where $f_1(^{40}\mathrm{K})n_p(^{40}\mathrm{K})$ gives the contribution (cpm) of the Compton scattering effect of ^{40}K to the ^{95}Zr + ^{95}Nb energy band, and D is a correction factor for the decay determined by the physical half-life of ^{95}Zr (parent of ^{95}Nb).

After determining the amount of ^{95}Nb, the amount of ^{95}Zr is obtained by the following equation.

$$^{95}\mathrm{Zr}(\mu\mu c) = {}^{95}\mathrm{Nb}(\mu\mu c)/k = {}^{95}\mathrm{Nb}(\mu\mu c)/2.2 \quad \dots\dots\dots\dots\dots\dots \quad (5)$$

The ^{137}Cs contained in the same sample caused some error in determining the $n_p(^{95}\mathrm{Zr} + {}^{95}\mathrm{Nb})$, since ^{137}Cs and ^{95}Zr + ^{95}Nb emitted similar energies. This error was taken into account, and, in the paper, the corrected values are listed.

For ^{103}Ru and ^{106}Ru Ruthenium-103 and ^{106}Rh (a daughter nuclide of ^{106}Ru) emit the same range of energies, and it was impossible to determine separately their activities*. Only the γ-disintegration rate (N(dpm)) of $^{103}Ru + ^{106}Ru$ at the energy band (0.43 ~ 0.57 MeV) is given by the following equation.

$$N(dpm) = \frac{n_p(^{103}Ru+^{106}Ru)-[f2(^{40}K)n_p(^{40}K)+f2(^{95}Zr+^{95}Nb)n_p(^{95}Zr+^{95}Nb)]}{\epsilon_p(^{103}Ru + ^{106}Ru)} \quad \ldots\ldots (6)$$

where $f_2(^{40}K)n_p(^{40}K)$ is the contribution of the Compton scattering effect of ^{40}K to the $^{103}Ru + ^{106}Ru$ energy band. and $f2(^{95}Zr + ^{95}Nb) \times n_p(^{95}Zr + ^{95}Nb)$ is the contribution of $^{95}Zr + ^{95}Nb$ to the same energy band.

For simplicity, it was assumed that either ^{103}Ru or ^{106}Ru emitted all the γ-rays from 0.43 to 0.57 MeV. The equations used for the calculation of ^{103}Ru and ^{106}Ru are as follows;

$$^{103}Ru(\mu\mu c) = \frac{N(dpm)}{\gamma(^{103}Ru)} \cdot \frac{D(^{103}Ru)}{2.22} \quad \ldots\ldots (7)$$

$$^{106}Ru(\mu\mu c) = \frac{N(dpm)}{\gamma(^{106}Rh)} \cdot \frac{D(^{106}Ru)}{2.22} \quad \ldots\ldots (8)$$

where $D(^{103}Ru)$ and $D(^{106}Ru)$ are the correction factors for the decays of ^{103}Ru and ^{106}Ru, respectively.

These two extreme values are presented in the paper.

2.4 Determination of ^{137}Cs

Diet and Feces Figure 1 shows a flow sheet of the procedure used for determination of ^{137}Cs. Cesium and strontium chlorides (30 mg each) were added as carriers to the ashed sample. The sample was treated with about 200 ml of (1:1) hydrochloric acid on a hot plate for about 30 min, and the insoluble materials were filtered off. The acid treatment was repeated with 50 to 100 ml of the hydrochloric acid solution, usually three times, until the solution did not show a yellow color. The solution was then evaporated, and 40 ml of concentrated nitric acid was added to the residue and evaporated again. After adding 40 ml of hydrochloric

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* It may be possible to separate ^{103}Ru from ^{106}Ru by following the physical decay of the sample if the radioactivities are high (see Fig. 7 in later section), but it needs time and labor, and forbids the subsequent separation of ^{90}Sr. Therefore, this was not taken in the paper.

acid, the solution was evaporated again. The residue was subsequently mixed with 500 ml of hot water, and the insoluble material was filtered off. After adding 100 ml of hot water to the insoluble material, the filtration was repeated. The residue was discarded.

All the filtrates were combined, and (1:1) ammonium hydroxide solution was added until the pH of the solution reached about 7. Next, 2M ammonium carbonate was added sufficiently to the solution.

The carbonate precipitate obtained was separated by filtration, and washed with 30 ~ 40 ml of water. The carbonate precipitation procedure described above was repeated. From the carbonate precipitates, ^{90}Sr was determined (see section 2.5).

For ^{137}Cs, the filtrates and washings were collected and mixed with concentrated nitric acid (about 1/5 of the filtrate volume). The solution was heated to 80 ~ 90°C. Then, addition of 10 ml of 6N phosphoric acid and 200 ml of 10% ammonium molybdate to the solution produced phospho-ammonium molybdate precipitate. In this way, cesium was quantitatively precipitated as phospho-cesium molybdate(3). The precipitate was filtered off, and washed with a small amount of dilute nitric acid. After drying and weighing, the γ-activity (0.58 ~ 0.75 MeV) of the precipitate was measured by the 100 channel pulse-height analyser.

Urine In the determination of ^{137}Cs in urine, there was a method proposed by Hanya, but in this study the method was modified as follows (Fig. 2).

The filtrate which was obtained from urine by the phosphate precipitation procedure was taken as sample solution. To the filtrate, concentrated nitric acid (about 1/5 of the filtrate volume) was added, and the solution was heated to 80 ~ 90°C. Subsequently, phospho-ammonium molybdate was precipitated by adding 10 ml of 6N phosphoric acid and 150 ml of 10% ammonium molybdate solution. The γ-activity of the precipitate was measured by the method described above.

2.5 Determination of ^{90}Sr

The amount of ^{90}Sr in diet, feces and urine was determined by the procedure shown in Fig. 3. The procedure was based on that of the Committee on Radioactivity Measurement(4) but was somewhat different.

Diet and Feces As stated above, ^{90}Sr was contained in the carbonate precipitate which was separated in the cource of ^{137}Cs determination. About one half of the precipitate was dissolved with nitric acid, and the solution was dried up on a water bath. After the residue was dissolved with 15.0 ml of water, 57 ml of fuming nitric acid (s.g.;1.52) was added by drops with adequate stirring under ice cooling. Then strontium crystalized as nitrate. The mixture was stood for a few hours with occasional stirring, and the strontium nitrate was separated by a G4 glass filter, and washed with 5 ~ 6 ml of fuming nitric acid (s.g.;1.42) and dissolved with 10.0 ml of water. The fuming nitric acid procedure was repeated with 38 ml of fuming nitric acid (s.g.;1.52), and strontium nitrate was dissolved again with about 10 ml of water. As a scavenger, 1 ml of ferric chloride (10 mg as Fe) was added to the solution, and heated to about 90°C. Subsequently, ferric hydroxide was precipitated by adding (1:1) ammonium hydroxide solution, and the ferric hydroxide was filtered off and discarded. To the filtrate, 2 ml of barium chloride solution (20 mg as Ba), 1 ml of 6N acetic acid and 2 ml of 6M ammonium acetate solutions were added. The mixed solution was heated to 90°C. Barium was precipitated as chromate by adding 1 ml of 0.3M sodium chromate solution. The barium chromate was filtered off and washed with 5 ~ 6 ml of acetate buffer solution (pH;5.3). The filtrate and washings were combined and heated to about 100°C. Then 15 ml of 10% sodium carbonate solution was poured with stirring. Strontium precipitated as carbonate. The precipitate was filtered off, washed with a small amount of water, and then dissolved with 1 ~ 2 ml of (1:1) hydrochloric acid. After adding yttrium carrier (10 mg as Y), the solution was stood for about two weeks.

The solution was, then, boiled, and yttrium was precipitated as hydroxide with (1:1) ammonium hydroxide solution. The time the precipitation was done was recorded. To the mixture, 2 or 3 drops of 30% hydrogen peroxide solution were added. After boiling the mixture again, the yttrium hydroxide was filtered off. The precipitate was washed with a small amount of water and dissolved with 1 or 2 ml of (1:1) hydrochloric acid. After heating the solution to about 100°C, 0.2 ml of the strontium carrier solution (2 mg as Sr) was added, and the yttrium hydroxide precipitation procedure was repeated.

The hydroxide was dissolved with 1 or 2 ml of (1:1) hydrochloric acid, and yttrium oxalate was precipitated by addition of 5 ml of saturated oxalic acid. The oxalate precipitate was mounted on a filter paper in a demountable glass filtering apparatus. It was washed with a small amount of water and then with alcohol. The β-activity of the oxalate precipitate was measured with a Libby counter (the background activity;0.7 ~ 1.0 cpm). The decay of the ^{90}Y in the precipitate was also measured to ascertain the radiochemical purity of the observed activity.

Urine About one half of the phosphate precipitate obtained in the co-precipitation procedure, was used as the sample for ^{90}Sr. The precipitate was ashed with concentrated nitric acid, and then the residue was dissolved with 15.0 ml of water. The amount of ^{90}Sr was determined by the same method for diet and feces.

2.6 Determination of Calcium

The amount of calcium in diet, urine and feces was determined by modifying A.O.A.C. method[5].

Diet and Feces About 0.1 g of the carbonate precipitate obtained in the ^{137}Cs determination was dissolved with a small amount of dilute hydrochloric acid, and diluted to 200 ml with water. Two milliliters of the solution was taken into a small centrifugal tube. Two milliliters of (1:4) acetic acid and 2 ml of saturated ammonium oxalate solution were then added. The pH of the solution was adjusted with ammonium hydroxide to 5.0, using methyl red as an indicator, and the solution was stood overnight in order to precipitate completely the calcium as the oxalate. The mixture was centrifuged at 3200 rpm for 15 min, and the supernatant solution was siphoned off. The oxalate precipitate was washed with 10 ml of (1:49) ammonium hydroxide solution, and the mixture was centrifuged again. The supernatant solution was siphoned off. This washing with ammonium hydroxide solution was repeated. The oxalate precipitate was dissolved with 2 ml of (1:4) sulfuric acid. The solution was titrated with a normal solution of 1/50N potassium permanganate at 60 ~ 70°C.

The titration was done four times for each determination and the average value was obtained.

Urine Two milliliters of urine was taken into a small centrifugal tube, and the calcium content was determined by the same method mentioned above.

2.7 Determination of Potassium

Diet and Feces The stable potassium content in diet and feces was calculated from the amount of ^{40}K measured by γ-spectrometry. In this calculation, the abundance ratio and specific activity of ^{40}K were taken as 0.0119% and 7.2×10^{-6} c/g, respectively.

Urine In this case, potassium was determined chemically. Twenty milliliters of urine was taken as a sample. First, wet ashing with nitric acid was made and then the residue was mixed with 15 ml of concentrated hydrochloric acid. The mixture was dried up. After the residue was dissolved with 25 ml of 1N hydrochloric acid, the solution was filtered and the filter paper was rinsed with water. The filtrate and rinsings were combined, and diluted to about 50 ml with water. To the solution, 10 ml of 37% aqueous solution of formaldehyde and 10 ml of potassium-free sodium hydroxide solution* were added, and moreover, 40 ml of sodium

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* Thirty grams of sodium hydroxide was dissolved with 200 ml of water. To the solution, 38 ml of sodium tetraphenyl borate solution was added with stirring. The mixed solution was diluted quantitatively to 250 ml. The solution was filtered by dry filter paper. In this way, potassium in the sodium hydroxide was eliminated.

tetraphenyl borate solution* was poured with stirring. The solution was diluted quantitatively to 200 ml with water, and filtered by dry filter paper. Fifty milliliters of the filtrate was taken and kept at about 30°C. An excess amount of sodium tetraphenyl borate in the filtrate was titrated with a normal solution of zephiramine** using titan yellow as an indicator.

The triple titration was carried out, and the average value was recorded.

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* Sodium tetraphenyl borate (12.2 g) was dissolved with 800 ml of water. After adding 20 ml of 0.5M alminium chloride solution, the solution was neutralized with 20% of sodium hydroxide solution, and diluted quantitatively to 1000 ml. The solution was filtered by dry filter paper. To the solution, 2 ml of sodium hydroxide solution was added with stirring. After one hour, the solution was filtered by dry filter paper.

** Six grams of tetradecyl-dimethyl benzyl ammonium chloride was dissolved with water, and diluted quantitatively to 1000 ml.

3. RESULTS AND DISCUSSION

3.1 Gamma-spectra of Samples

Typical γ-spectra of diet, urine, and feces are illustrated in Figs. 4, 5 and 6, respectively. In general, it was found that ashed diet and feces had contained considerable amounts of γ-emitters, whereas the precipitates of urine had not contained detectable amounts of γ-emitters, except ^{137}Cs and ^{40}K (these nuclides existed in the filtrates).

3.2 Levels of ^{40}K, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru

The average values and ranges of ^{40}K, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru in diet, urine, and feces of the Subjects II ~ V, are summarized in Table 3. The values for the individuals are shown in Table 4.

In general, the radioactivity of ^{95}Nb in diet was highest in all fission nuclides, reaching about 60 $\mu\mu$c/day/person in February 1963. However, it may be suspected from the figures shown in Tables 3 and 4 that the level of ^{95}Nb was lower than that of ^{106}Ru in some cases, but this was unlikely, since in this study, the mixtures of ^{103}Ru and ^{106}Ru were measured and the amount of ^{106}Ru involved was perhaps smaller than that of ^{103}Ru, as shown by the patterns of physical decay of $^{103}Ru + ^{106}Ru$ in a fecal sample (Fig. 7).

It was noticed that the amounts of γ-emitters in feces varied considerably among the subjects. The highest activity was observed in the ashed sample from Subject I, collected on November 12 ~ 16, 1962. In this case, the amount of ^{95}Zr reached 110 $\mu\mu$c per day and that of ^{95}Nb exceeded 200 $\mu\mu$c per day.

Radio-autographic test of the sample was carried out, using X-ray film (15 × 25 cm), to seek for any large radioactive particles which might exist in it. The ashes of the diet collected in the same period were also examined. In the case of the fecal sample, one strong spot and several faint spots were observed on the film after one week exposure, but for the ashed diet sample, one or two very faint spots were only observed.

To test the solubility, the fecal ash was treated with (1:1) hydrochloric acid. The γ-activity of $^{95}Zr + ^{95}Nb$ did not leach out with the acid, but remained with insoluble materials (the main portion was carbon, which was due to insufficient ashing). The insolubility of $^{95}Zr + ^{95}Nb$ may be shown again by the low level activity in urine and the high level in feces.

3.3 Levels of ^{90}Sr and ^{137}Cs

Table 5 gives the average level and range of ^{90}Sr and ^{137}Cs

for the Subjects II ~ V in $\mu\mu c$/day/person, as well as in strontium unit or in cesium unit. Table 6 shows the levels for each subject.

The levels of ^{90}Sr in diet varied between 7.0 and 13 $\mu\mu c$/day/person, and those of ^{137}Cs, between 13 and 35 $\mu\mu c$/day/person. However, about 1 $\mu\mu c$ of ^{90}Sr was always found in 24-hr urine samples, irrespective of the content in the diet. In contrast with ^{90}Sr, the amount of ^{137}Cs in the same specimens varied between 10 and 31 $\mu\mu c$ corresponding to the contents in the diet. As illustrated in Table 5, it was noticed that the ratio of ^{137}Cs to ^{90}Sr in diet did not much deviate from twice during the period examined.

As the materials of the diet in this experiment were mainly produced in and arouno Tokai village, the levels of $^{95}Zr + ^{95}Nb$, ^{90}Sr and ^{137}Cs in the diet were compared with the concentrations in the air at Tokai. The concentrations in the air were determined by Kasai(6) from the dust collected on filter paper with an air sampler. Figures 8 and 9 show the activity levels of $^{95}Zr + ^{95}Nb$ and ^{137}Cs in the diet and in the air, indicating the close correlation between the two.

The correlation in $^{95}Zr + ^{95}Nb$ may suggest that the levels of $^{95}Zr + ^{95}Nb$ in the diet were not determined by the cumulative deposition on the ground (or absorption from root), but by short-term deposit of the isotopes on the vegetables. With reference to ^{137}Cs, the situation will be roughly interpreted as follows. Deposition of ^{137}Cs on the surfaces of vegetables (taken by the subjects directly) and weeds (eaten by cows and domestic animals) would be proportional, in a first approximation, to the levels of monthly deposit. The levels of ^{137}Cs in milk or dairy products were perhaps proportional to the content in the weeds. Considering the subjects took daily 360 ml of milk which might be the main source of ^{137}Cs (see Table 5 and Reference (7)), the diet would contain ^{137}Cs, after all, in proportion to the levels of the nuclides in the air, even if some vegetation and cereals might absorb the nuclides from the ground and give some contributions to the contents in the total diet.

3.4 Weight and Volume of Samples and Contents of Calcium and Potassium

Diet The daily weight of the diet and the contents of calcium and potassium are summarized in Table 7. Figure 10 shows the frequency distribution of the weight of the ashed diet. It was found that the average weight of the ashed diet was about 19 g. This figure corresponds to about 490 g of diet in the dry state and 2200 g in the wet state.

The amounts of calcium and potassium were 0.56 g and 1.73 g/day/person, respectively. The diet in the experiment was probably that of urban type in Japan, judging from the quality and amount. However, the content of calcium (0.56 g) differed from the average value of the urban diets (0.40 g). This discrepancy may be explained by the fact that the subjects received two bottles (360 ml) of milk in addition to the usual meals, whereas typical Japanese urban diets contain only 45 g of milk and dairy products.

Urine Table 8 presents the volume of urine, the weight of phosphate precipitate and the contents of calcium and potassium per day. Figure 11 gives graphically the frequency distribution of daily volume and the weight of the phosphate in the Subjects II ~ V. On the other hand, Figure 12 shows those for each subject.

The average daily volume of urine for the individuals was 950 to 1300 ml, and the standard deviation was 30 to 40 per cent. The weights of the phosphate precipitates of calcium and magnesium were 1.0 to 1.5 g per day, and the standard deviation was 20 to 40 per cent.

The correlation coefficient between the urinary volume and the weight of the phosphate was calculated, and it was only 0.025 (Fig. 13).

Furthermore, the correlation between the urine volume and the contents of calcium and potassium is illustrated in Figs. 14 and 15. The correlation coefficients for the calcium and for the potassium were 0.28 and 0.35, respectively.

Figure 16 gives the correlation between the amounts of phosphate and calcium and potassium contents. The correlation coefficient for the latter was 0.064, and that for the former, 0.59.

Feces Table 9 shows the weight of the ashes per day as well as the contents of calcium and potassium per week, for each subject. The frequency distribution of the ash weights of all feces from the Subjects II ~ V is shown in Fig. 18, and that for each individual, in Fig. 19.

As may be seen from Table 9, the daily weights of feces varied considerably. Consequently, the fecal data must be treated carefully when they are used for the evaluation of internal exposure.

The results obtained in the present experiment may be used in the studies of the metabolism of radioactive nuclides in man. These studies of metabolism will be described in other papers by one of the authors.

Acknowledgments

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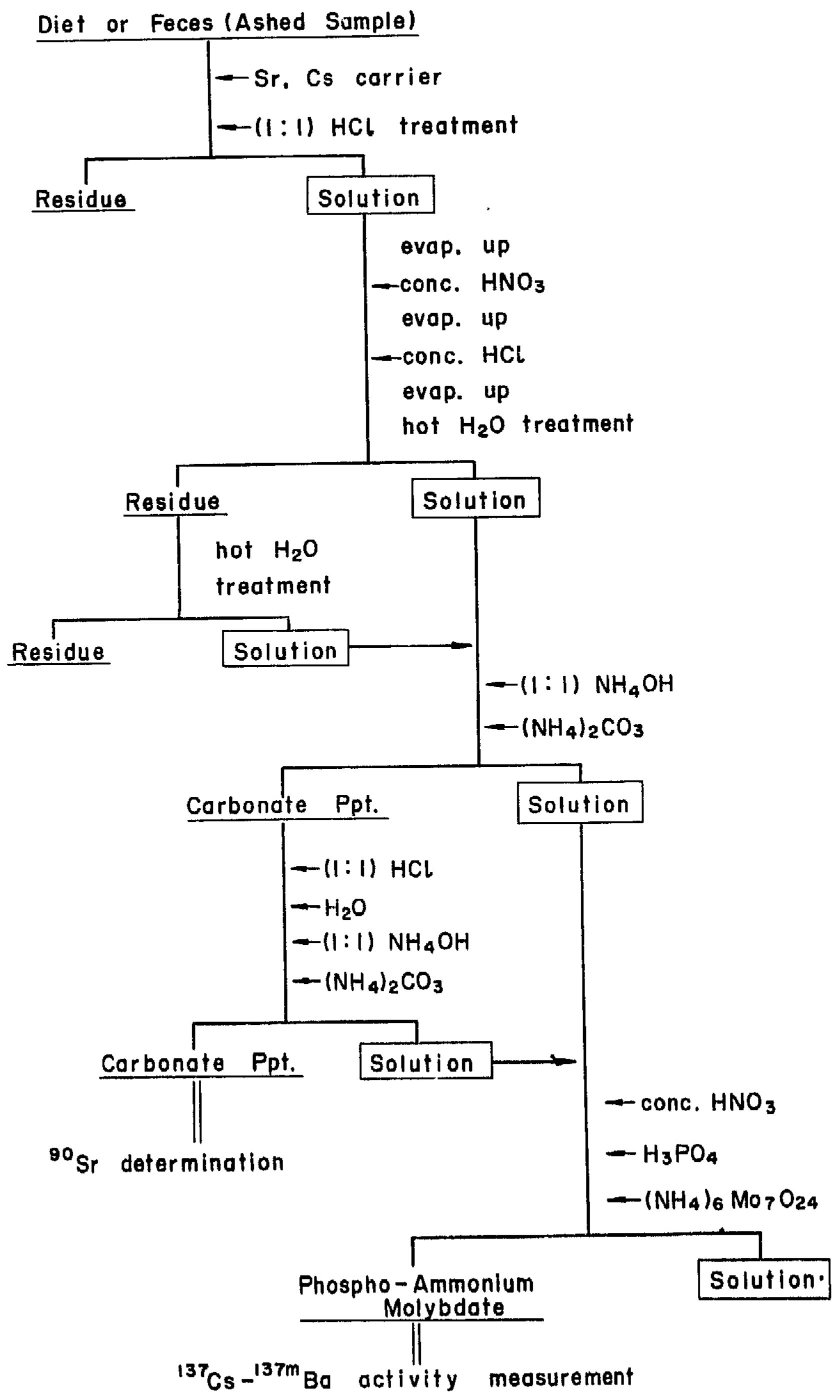


Fig. 1. Determination procedure of ^{137}Cs in diet and feces.

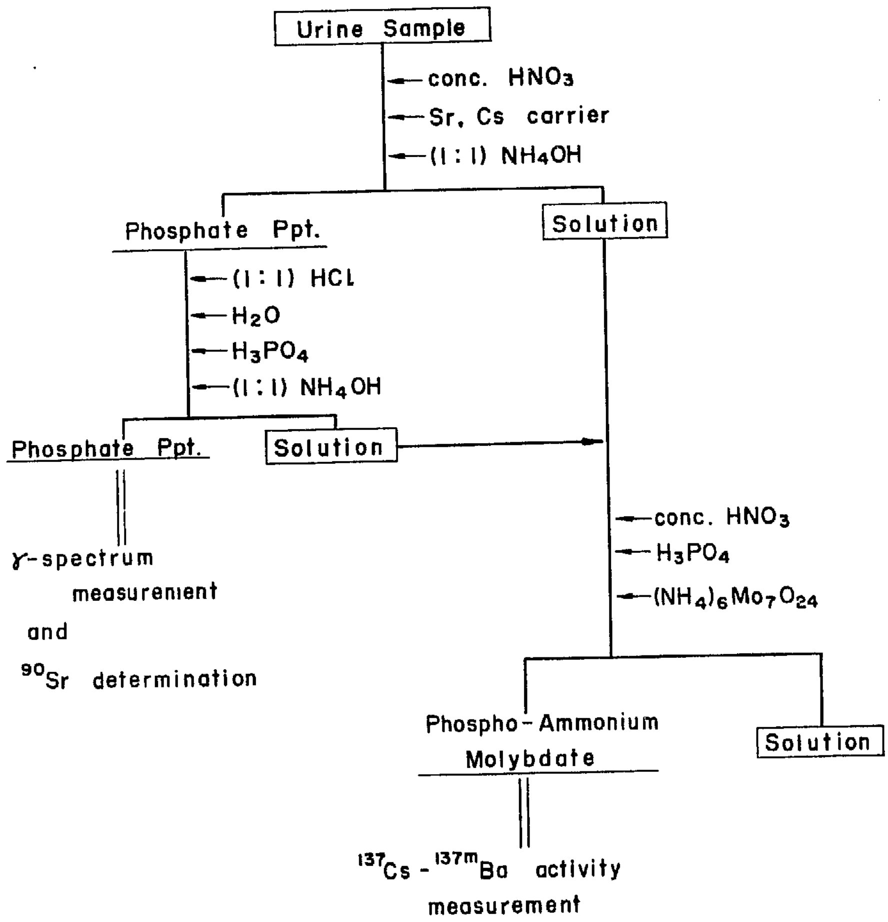


Fig. 2. Determination procedure of ^{137}Cs in urine.

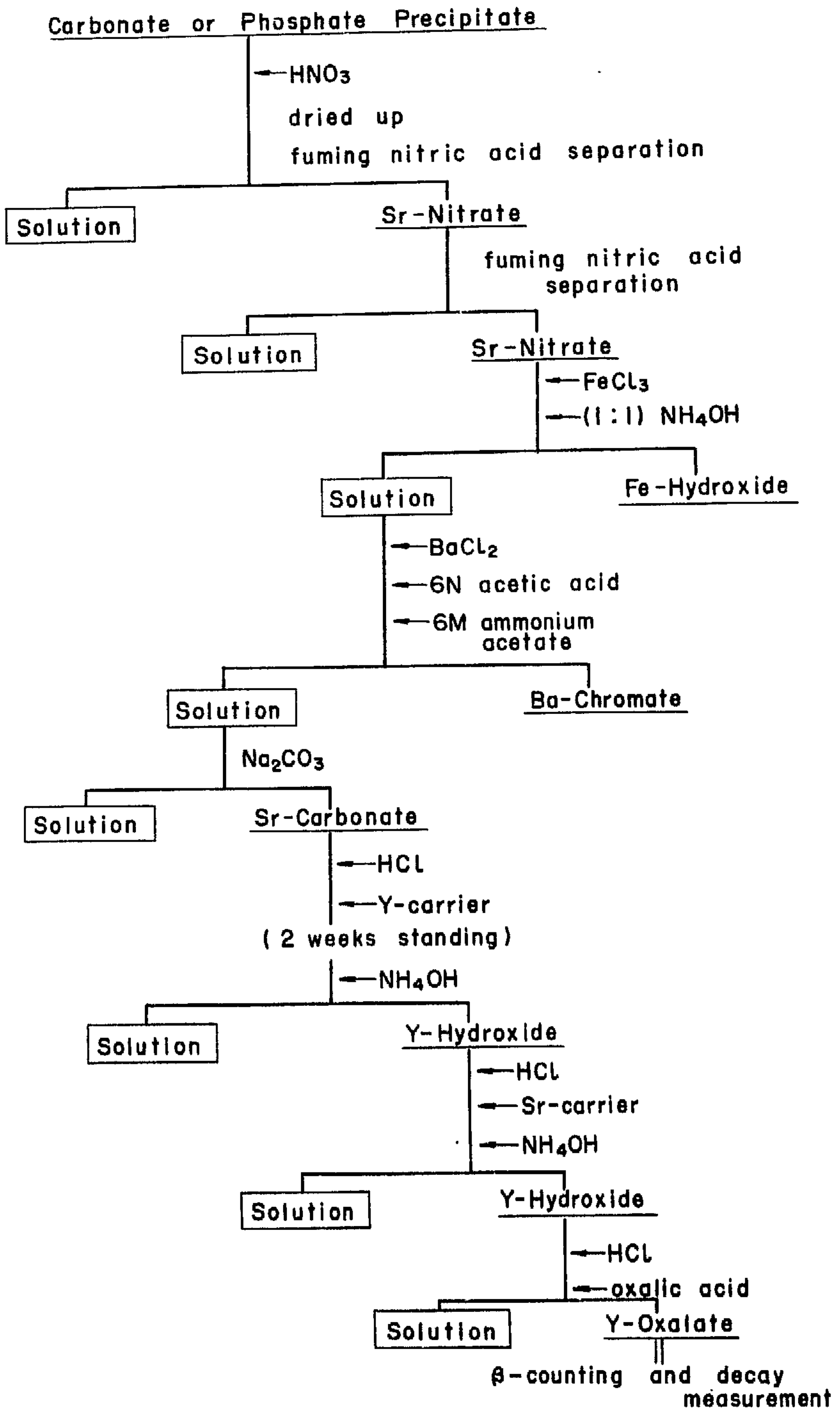
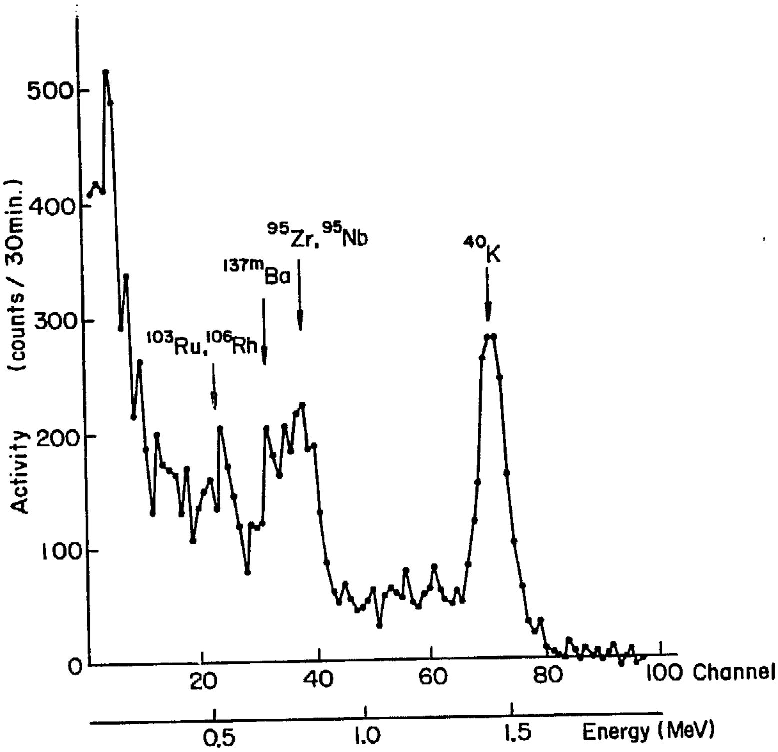


Fig. 3. Determination procedure of ^{90}Sr in diet, feces and urine.

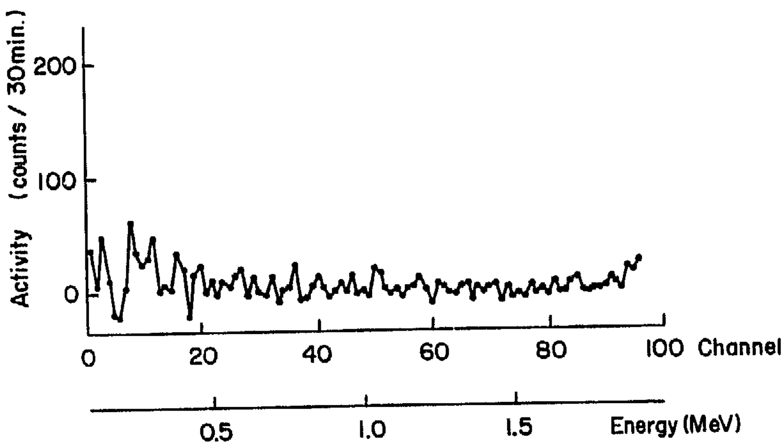


Sample : Diet (I person - 5 days)

Sampling date : 12~16, Nov., 1962

Counting date : 26, Nov., 1962

Fig. 4. Gamma-spectrum of ashed diet.



Sample : Urine (4 persons - 5 days)

Sampling date : 12~16, Nov., 1962

Counting date : 28, Nov., 1962

Fig. 5. Gamma-spectrum of phosphate precipitate of urine.

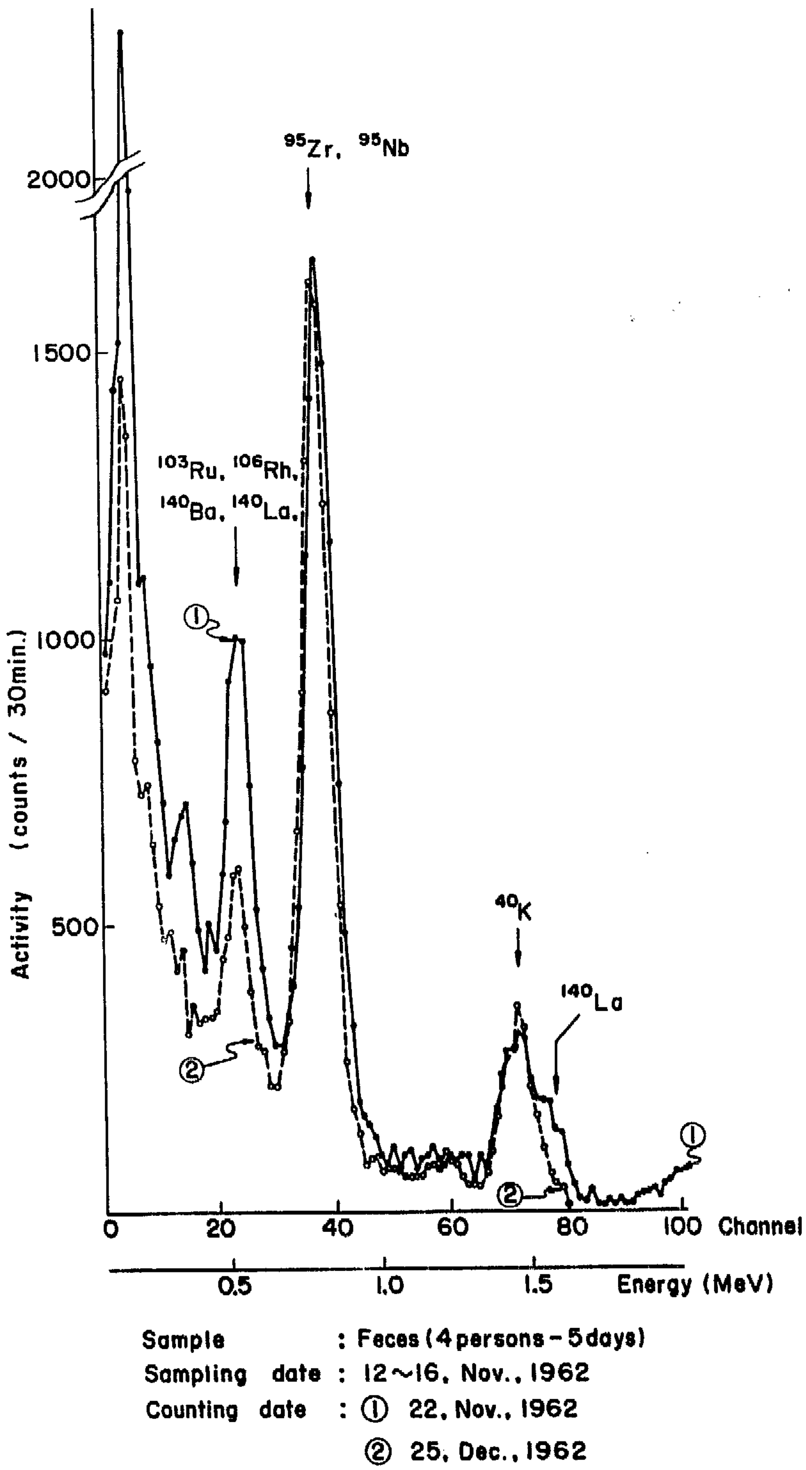


Fig. 6. Gamma-spectrum of ashed feces.

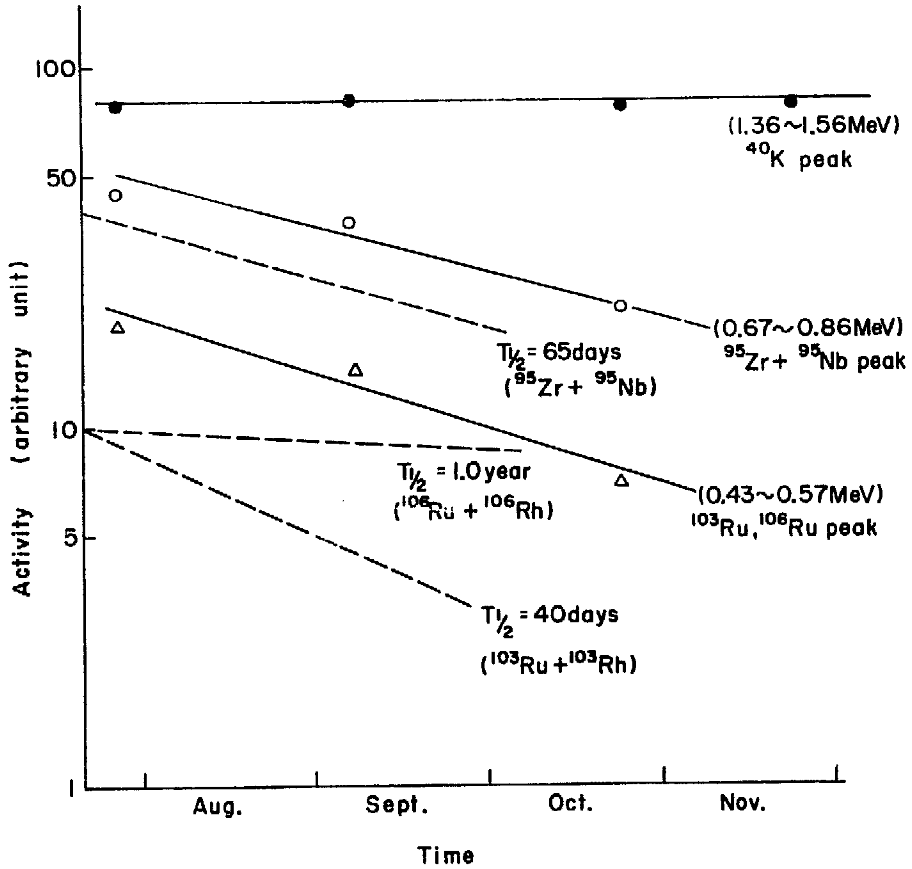


Fig. 7. Radioactive decay of fecal ashes, collected May 21 ~ 25, 1962.

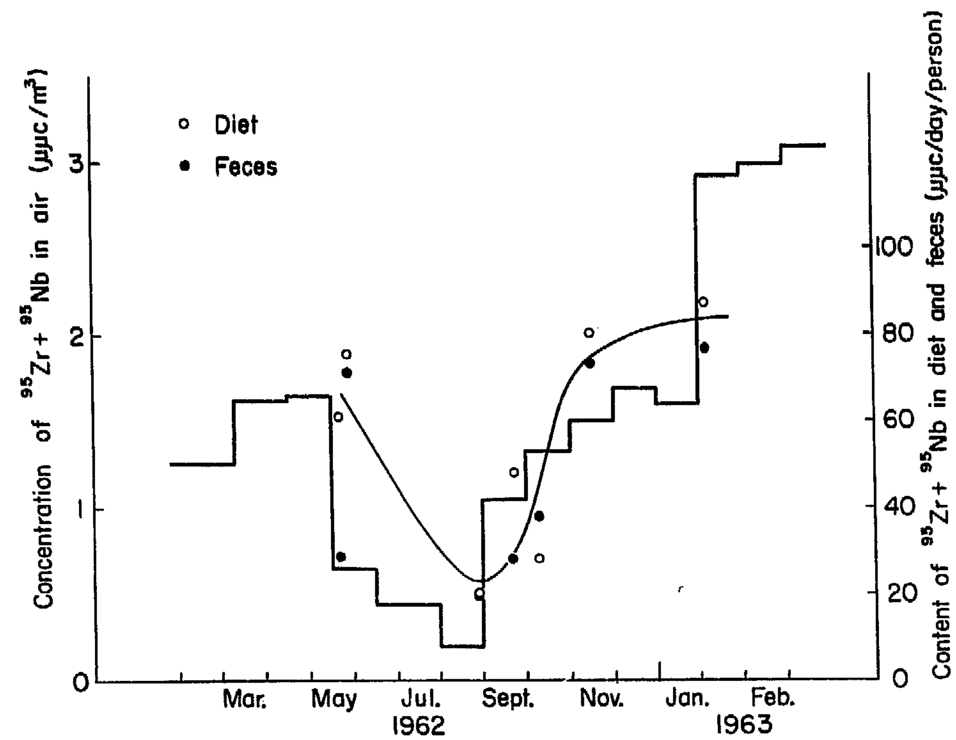


Fig. 8. Comparison of $^{95}Zr + ^{95}Nb$ level in diet and feces with that in air.

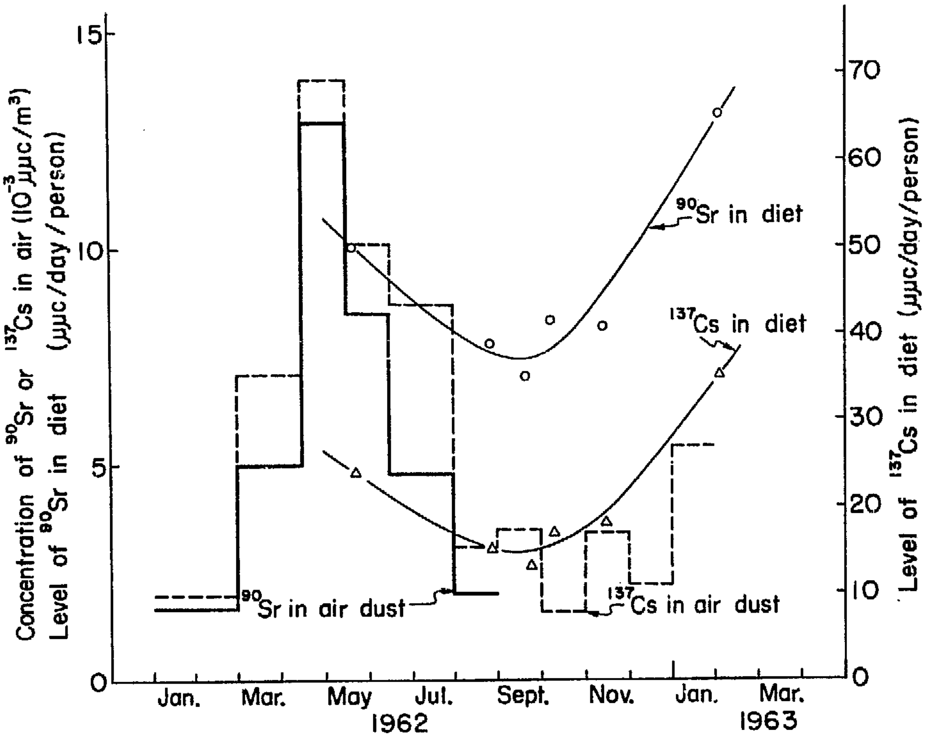


Fig. 9. Change of ^{90}Sr and ^{137}Cs levels in diet and air.

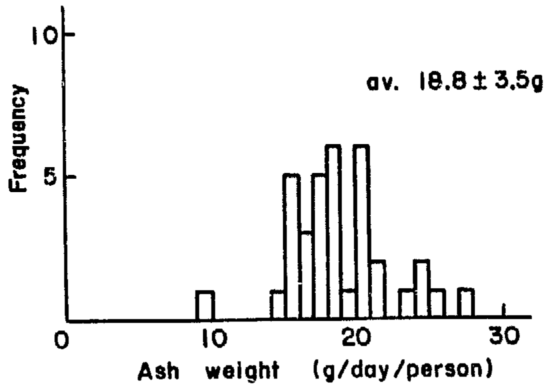


Fig. 10. Frequency distribution of dietary ash weight.

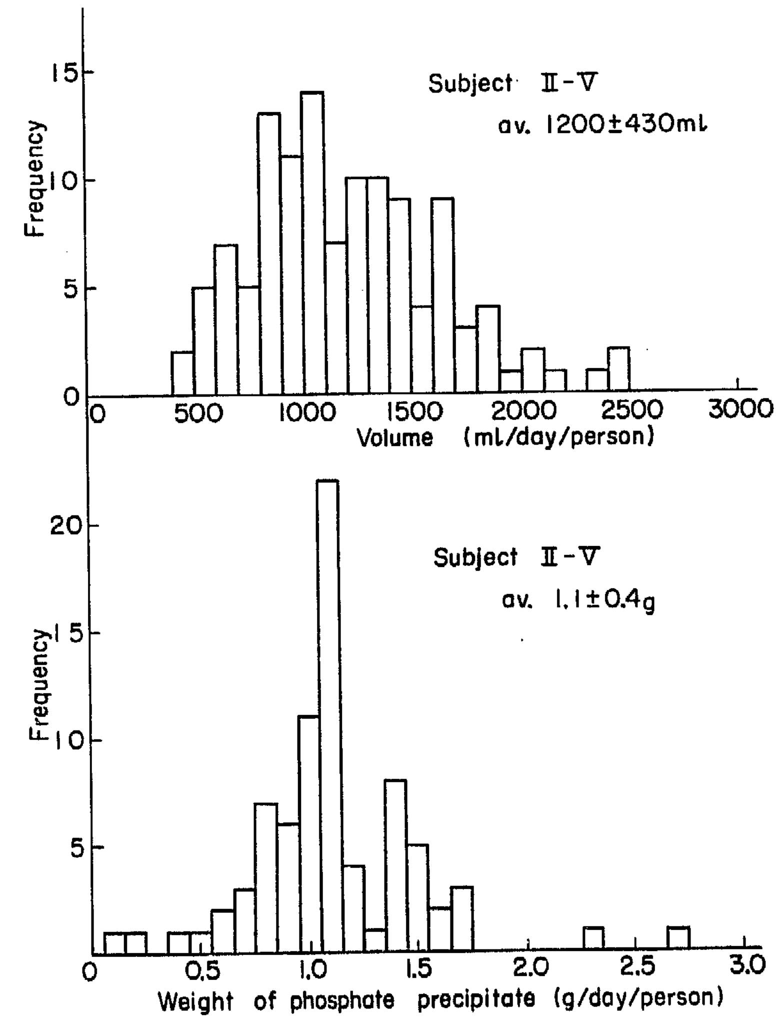


Fig. 11. Frequency distribution of urinary volume and weight of phosphate precipitate.

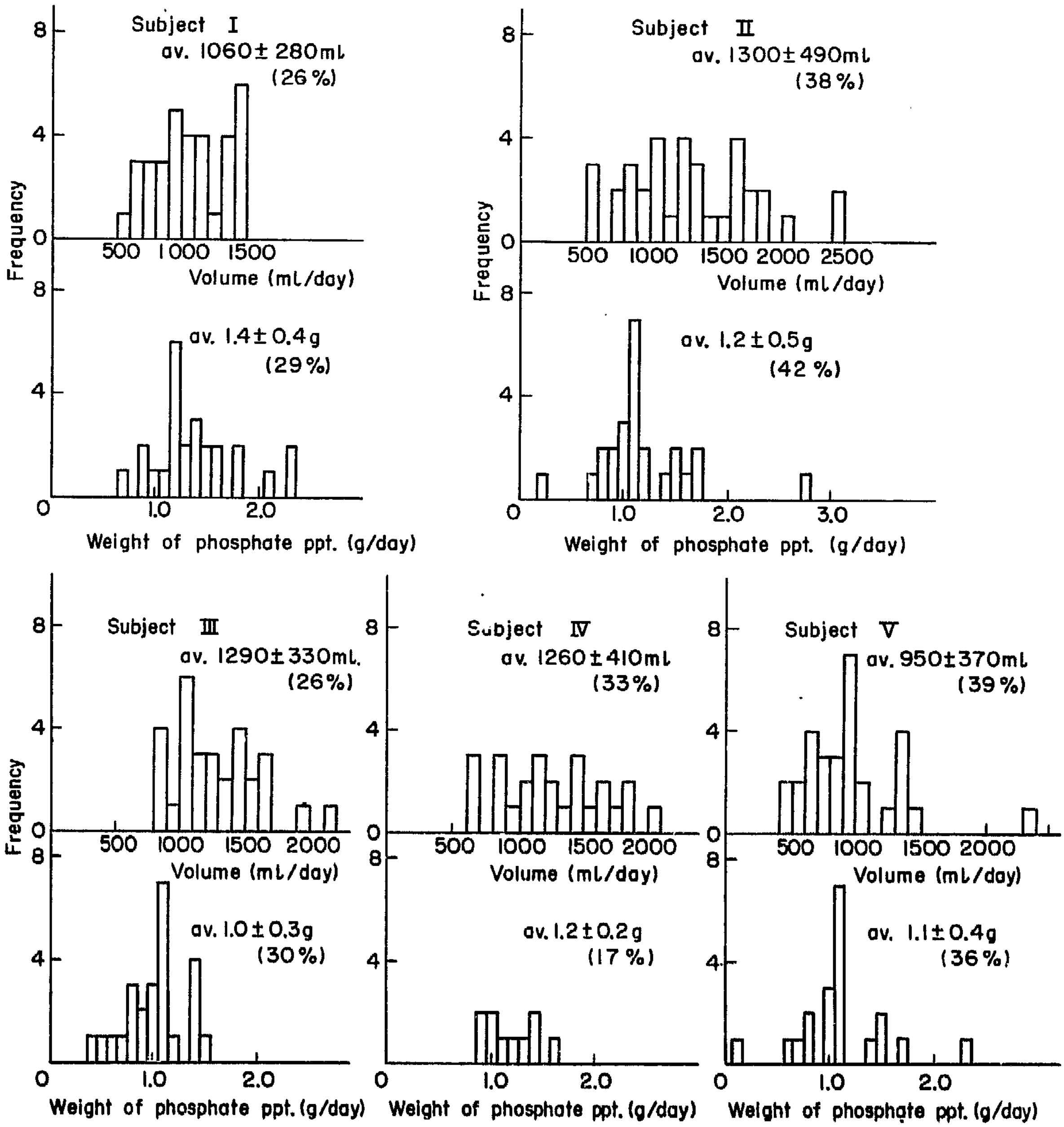


Fig. 12. Frequency distribution of urinary volume and weight of the phosphate precipitate for each subject.

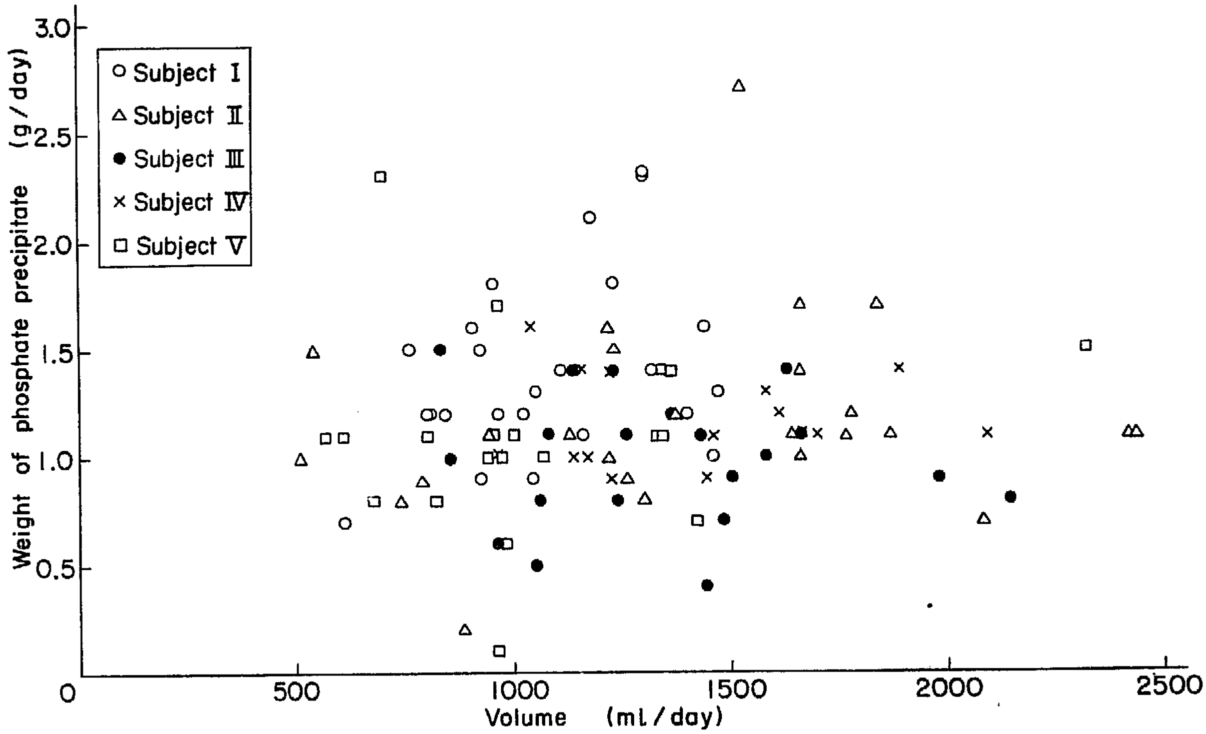


Fig. 13. Correlation between volume and phosphate precipitate in urine.

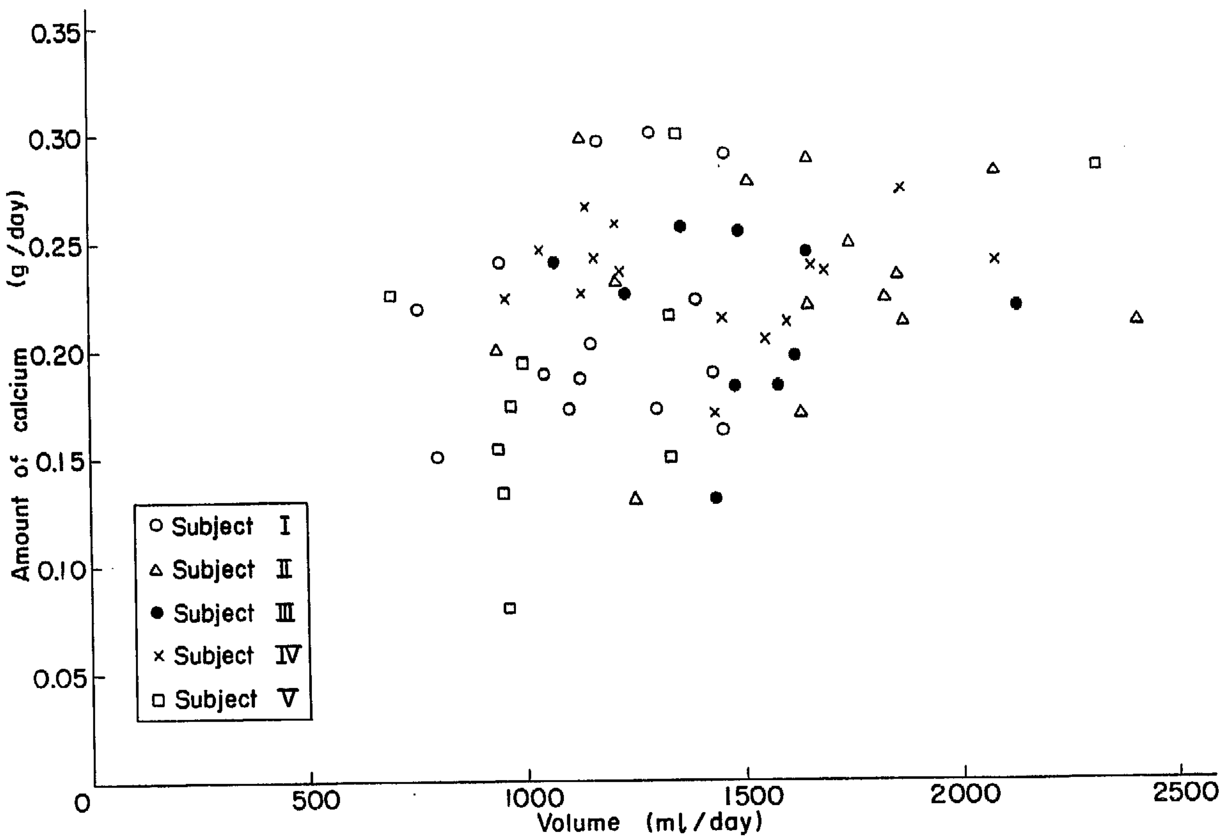


Fig. 14. Correlation between volume and calcium content in urine.

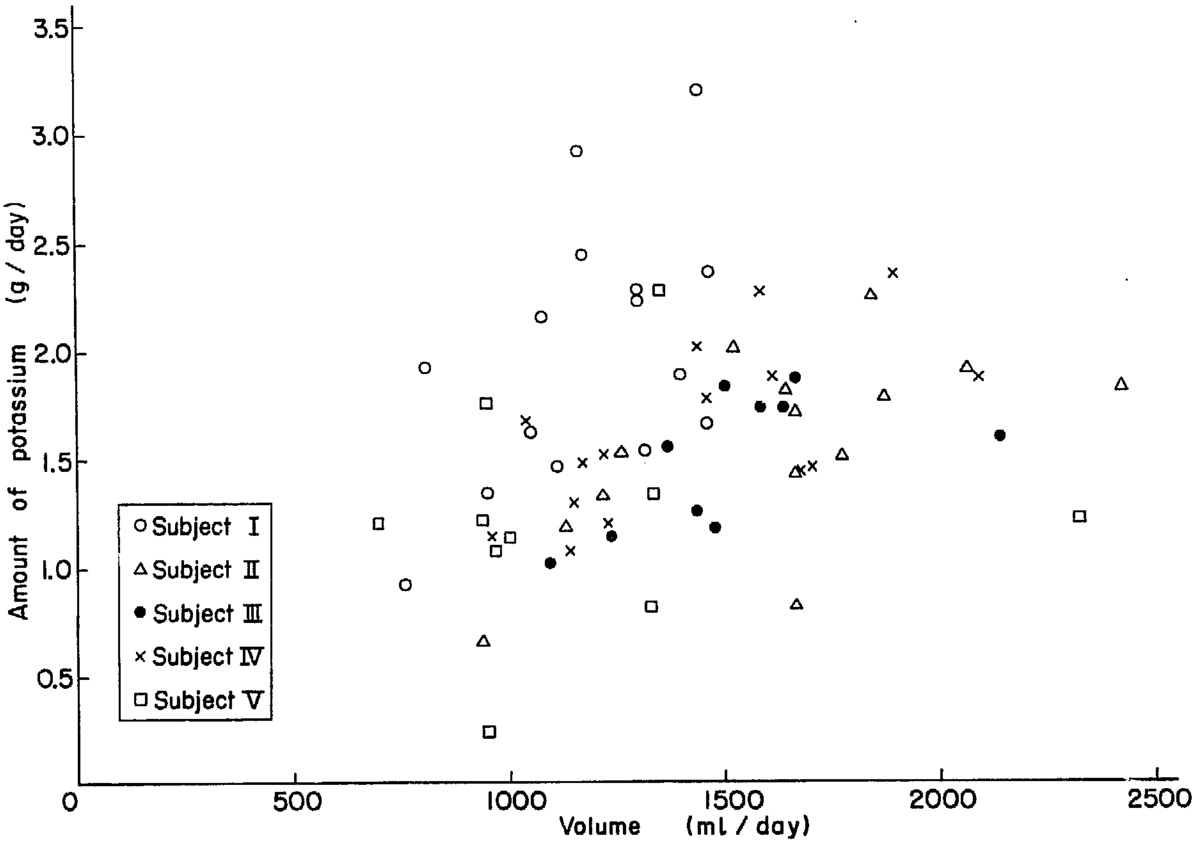


Fig. 15. Correlation between volume and potassium content in urine.

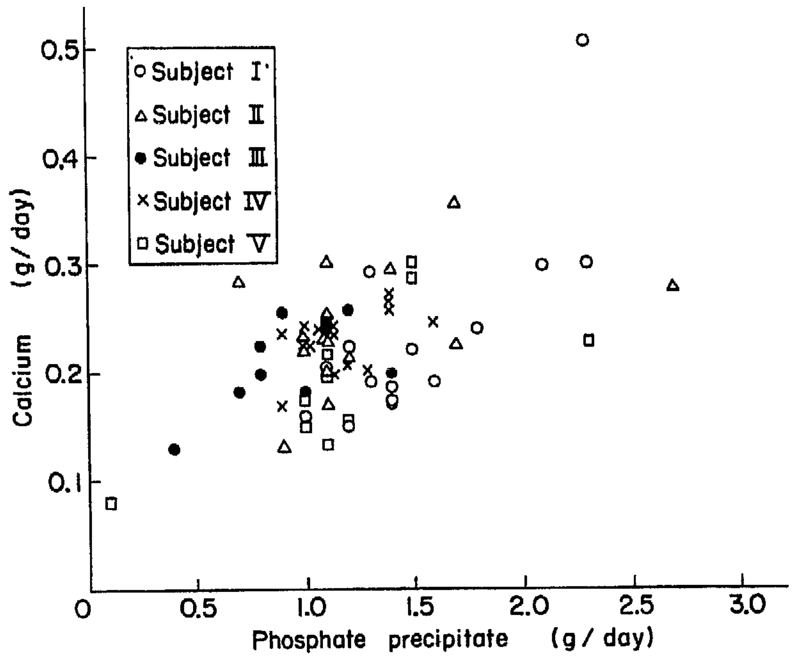


Fig. 16. Correlation between phosphate precipitate and calcium in urine.

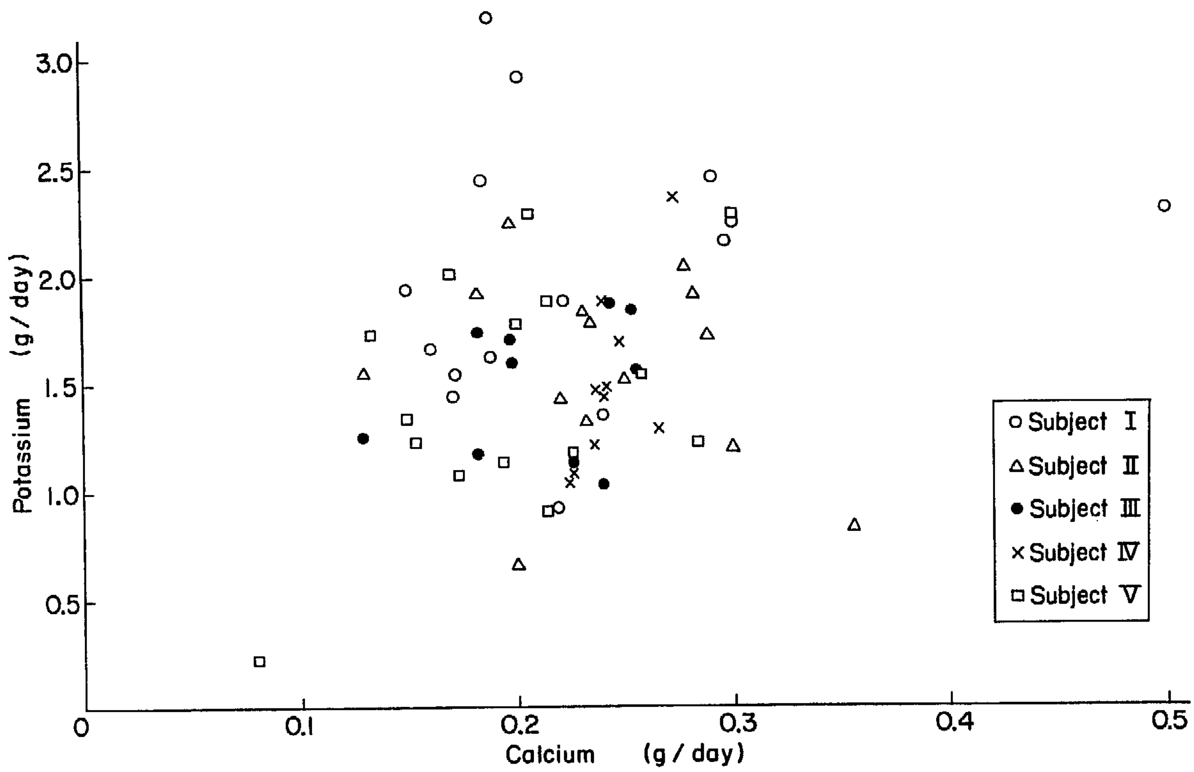


Fig. 17. Correlation between calcium content and potassium content in urine.

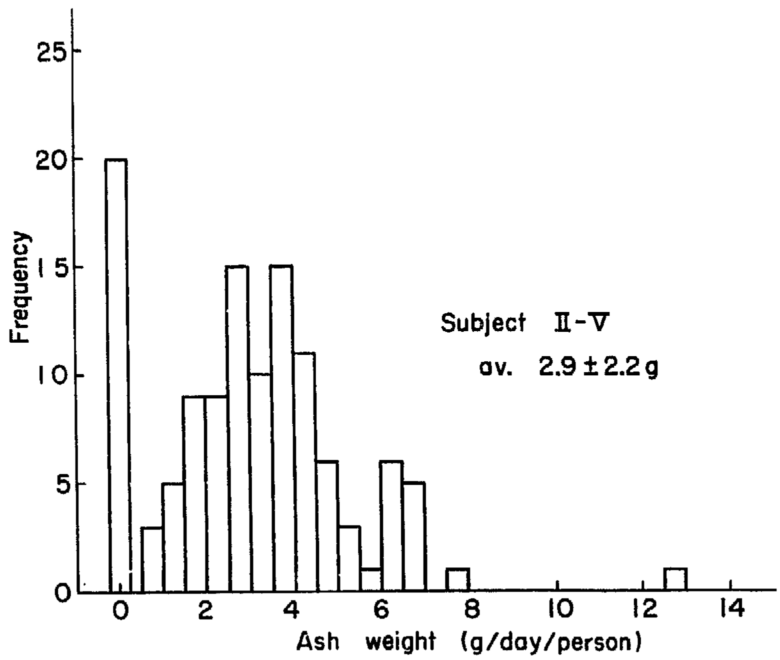


Fig. 18. Frequency distribution of fecal ash wei..ıt.

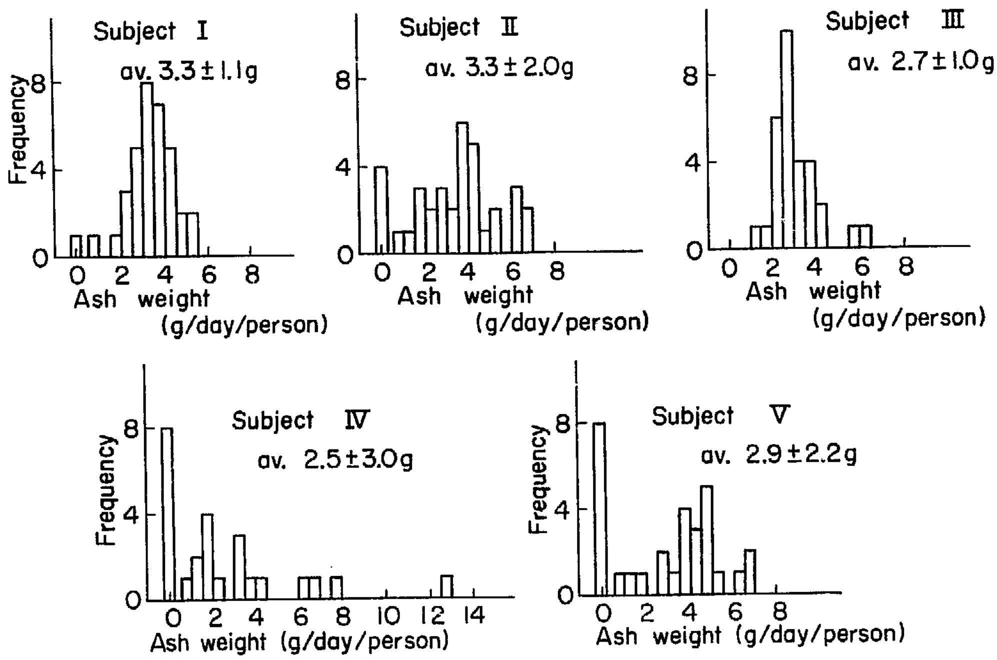


Fig. 19. Frequency distribution of fecal ash weight.

Table 1. Age, Height, and Weight of the Subjects

Subject	Age	Height (cm)	Weight (kg)
I	39	166	57
II	34	165	56
III	30	160	48
IV	24	161	59
V	23	166	55

Table 2. Energy Band, Peak Efficiency and Detectable Limit

Sample Size (cm)	Distance (cm)	Nuclide	Energy Band (MeV)	Peak Efficiency	Detectable Limit ($\mu\mu c$/day/person)
7 × 7 × 3	4.5	^{40}K	1.36 ~ 1.56	0.0364	40
7 × 7 × 3	4.5	$^{95}Zr + ^{95}Nb$	0.67 ~ 0.86	0.0610	2(^{95}Zr), 3.5(^{95}Nb)
7 × 7 × 3	4.5	$^{103}Ru + ^{106}Ru$	0.43 ~ 0.57	0.0825	6(^{103}Ru), 20(^{106}Ru)
7 × 7 × 1	2.5	^{40}K	1.36 ~ 1.56	0.0537	17
7 × 7 × 1	2.5	$^{95}Zr + ^{95}Nb$	0.67 ~ 0.86	0.0994	1(^{95}Zr), 2(^{95}Nb)
7 × 7 × 1	2.5	$^{103}Ru + ^{106}Ru$	0.43 ~ 0.57	0.145	3(^{103}Ru), 10(^{106}Ru)
7 × 7 × 0.5	2.5	^{137}Cs	0.58 ~ 0.75	0.130	3

Table 3. Average Levels and Range of ^{40}K, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru in Diet, Urine and Feces

($\mu\mu c$/day/person)

Nuclide	Sample	1962						1963
		May 21-25	*May 28 - June 1	Aug. 27-31	*Sep. 17-21	Oct. 8 12	*Nov. 12-16	*Feb. 4-8
^{40}K	Diet	1200	1600	1400	1400	1300	1400	2200
	Urine	—— (—)	—— (—)	—— (—)	—— (—)	1200 (1140~1330)	1200 (920~1380)	1500 (1270~1620)
	Feces	350 (—)	370 (340~410)	290 (200~380)	260 (200~410)	280 (200~410)	340 (320~370)	410 (300~680)
^{95}Zr	Diet	19	24	6	15	8.6	25	27
	Urine	—	—	—	—	—	—	—
	Feces	9	23 (15~35)	6 (3~10)	9 (7~12)	12 (6~20)	23 (15~30)	24 (18~37)
^{95}Nb	Diet	42	52	14	33	19	55	59
	Urine	—	—	—	—	—	—	—
	Feces	19	49 (34~74)	14 (8~20)	20 (15~26)	27 (14~43)	50 (33~66)	53 (39~81)
^{103}Ru	Diet	28	45	—	—	12	32	16
	Urine	—	—	—	—	—	—	—
	Feces	14	— (~14)	— (~7)	13 (11~14)	9 (3~12)	24 (16~32)	16 (10~20)
^{106}Ru	Diet	39	66	—	—	37	100	50
	Urine	—	—	—	—	—	—	—
	Feces	21	— (~24)	— (~24)	24 (22~27)	25 (7~32)	49 (33~63)	42 (30~63)

* during these periods, figures show mean value and range (indicated in parenthesis) of excreta from 3 persons, but during other periods, from 4 persons.

—— :below the detection limit

— :not measured

In the fecal sample collected in Nov. 12 ~ 16, 1962, 26 $\mu\mu c$/day/person of ^{140}Ba and ^{140}La were found.

Table 4. Levels of ^{40}K, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru in Urine and Feces with Individuals

($\mu\mu$c/day/person)

Subject	Nuclide	Sample	1962 May 21-25	May June 28-1	Aug. 27-31	Sept. 17-21	Oct. 8-12	Nov. 12-16	1963 Feb. 4-8
I	^{40}K	Urine	—	—	—	—	1740	1570	1800
	^{40}K	Feces	—	470	240	280	430	580	680
	^{95}Zr	"	—	21	3	5	22	110	41
	^{95}Nb	"	—	48	8	12	48	230	91
	^{103}Ru	"	—	15	14	11	25	35	26
	^{106}Ru	"	—	25	13	12	68	70	84
II	^{40}K	Urine	—	—	—	—	1140	1380	1500
	^{40}K	Feces	—	340	210	200	230	370	380
	^{95}Zr	"	—	15	3	9	11	25	18
	^{95}Nb	"	—	34	8	19	23	53	39
	^{103}Ru	"	—	——	——	13	11	24	10
	^{106}Ru	"	—	——	——	14	29	49	33
III	^{40}K	Urine	—	—	—	—	1280	—	1270
	^{40}K	Feces	—	370	380	300	400	—	300
	^{95}Zr	"	—	35	9	7	20	—	37
	^{95}Nb	"	—	74	20	15	43	—	81
	^{103}Ru	"	—	14	7	11	12	—	20
	^{106}Ru	"	—	24	24	22	32	—	63
IV	^{40}K	Urine	—	—	—	—	1330	1230	1620
	^{40}K	Feces	—	—	200	—	200	340	570
	^{95}Zr	"	—	—	5	—	12	30	18
	^{95}Nb	"	—	—	11	—	26	66	39
	^{103}Ru	"	—	—	——	—	11	32	19
	^{106}Ru	"	—	—	——	—	30	63	30
V	^{40}K	Urine	—	—	—	—	1380	920	—
	^{40}K	Feces	—	410	370	280	270	310	—
	^{95}Zr	"	—	18	6	12	6	15	—
	^{95}Nb	"	—	39	14	26	14	33	—
	^{103}Ru	"	—	14	——	14	3	16	—
	^{106}Ru	"	—	18	——	27	7	33	—

— : not measured.

—— : below the detection limit.

In the fecal samples collected in Nov. 12~16, 1962, 29 $\mu\mu$c (Subject I), 30 $\mu\mu$c (Subject II), 25 $\mu\mu$c (Subject IV), 24 $\mu\mu$c (Subject V) per day per person of ^{140}La were found.

Table 5. Average Level and Range of ^{90}Sr and ^{137}Cs in Diet, Urine and Feces

($\mu\mu c$/day/person)
S.U., C.U.

Nuclide	Sample	1962 May 21–25	1962 *May June 28–1	1962 Aug. 27–31	1962 *Sep. 17–21	1962 Oct. 8–12	1962 *Nov. 12–16	*1963 Feb. 4–8
^{90}Sr	Diet	10 [15.6]	❨6.1❩	7.8 [16.2]	7.0 [10.6]	8.3 [16.0]	8.1 [13.5]	13 [24.5]
	Urine	—	0.9 (0.6~1.0)	1.0 (0.8~1.2)	1.1 (1.0~1.1)	1.4 (1.2~1.5) [5.8]	1.0 (1.0~1.0) [4.6]	1.2 (1.0~1.4) [6.0]
	Feces	6.4 (5.2~8.4) [21.4]	6.9 (4.3~8.8) [32.8]	4.6 (3.4~5.3) [19.2]	4.1 (3.2~5.3) [14.6]	5.5 (4.9~9.9) [21.2]	6.7 (5.1~7.9) [18.1]	8.0 (5.6~9.3) [18.6]
^{137}Cs	Diet	24 [17.1]	❨10❩	15 [9.1]	13 [7.9]	17 [11.2]	18 [11.1]	35 [13.6]
	Urine	—	11 (10~13)	18 (15~21)	14 (11~16)	17 (12~20) [11.8]	22 (18~24) [16]	31 (25~36) [18]
	Feces	1.4 [3.4]	3.9	6.0 [17.7]	2.9 [9.7]	2.6 [7.9]	3.3 [8.3]	5.1 [13]

Figures show mean value and range (shown in parenthesis ( )) in $\mu\mu c$/day/person, figures in parenthesis [ ] show mean value in S.U. or C.U.

* during the periods, figures show mean value and range of excreta from 3 persons (other periods, 4 persons)

— : not measured.

❨ ❩ without milk

Table 6.. Levels of ^{90}Sr and ^{137}Cs in Urine and Feces

Subject	Nuclide	Sample	Unit	1962 May 21-25	1962 May June 28-1	1962 Aug. 27-31	1962 Sept. 17-21	1962 Oct. 8-12	1962 Nov. 12-16	1963 Feb. 4-8
I	^{90}Sr	Urine	μμc	—	0.9	0.5	0.6	1.2	1.0	1.7
			S.U.	—	—	—	—	4.5	4.8	7.5
		Feces	μμc	6.8	5.0	3.4	3.5	9.7	7.0	7.4
			S.U.	17.6	29.4	15.9	18.0	33.9	23.2	31.3
	^{137}Cs	Urine	μμc	—	14	18	11	19	26	35
			C.U.	—	—	—	—	9.3	14.1	16.6
II	^{90}Sr	Urine	μμc	—	0.6	1.2	1.0	1.4	1.0	1.0
			S.U.	—	—	—	—	4.9	4.2	5.1
		Feces	μμc	8.4	4.3	5.7	3.2	9.9	7.2	9.0
			S.U.	19.5	20.6	16.5	11.2	22.8	16.9	16.8
	^{137}Cs	Urine	μμc	—	13	19	16	20	24	33
			C.U.	—	—	—	—	15.0	14.8	18.8
III	^{90}Sr	Urine	μμc	—	1.0	1.0	1.1	1.2	—	1.1
			S.U.	—	—	—	—	5.1	—	5.8
		Feces	μμc	6.4	7.7	3.4	3.9	4.9	—	5.6
			S.U.	25.8	33.7	18.9	15.0	25.8	—	18.4
	^{137}Cs	Urine	μμc	—	10	15	14	17	—	25
			C.U.	—	—	—	—	11.3	—	16.8
IV	^{90}Sr	Urine	μμc	—	—	0.9	—	1.5	1.0	1.4
			S.U.	—	—		—	6.1	4.1	6.7
		Feces	μμc	5.2	—	4.3	—	6.3	7.9	9.3
			S.U.	22.2	—	15.2	—	22.8	16.2	20.6
	^{137}Cs	Urine	μμc	—	—	21	—	19	24	36
			C.U.	—	—		—	12.2	16.7	18.9
V	^{90}Sr	Urine	μμc	—	1.0	0.8	1.1	1.3	1.0	—
			S.U.	—	—		—	6.3	5.7	—
		Feces	μμc	5.4	8.8	4.8	5.3	4.8	5.1	—
			S.U.	19.1	44.5	32.9	18.3	34.7	26.6	—
	^{137}Cs	Urine	μμc	—	10	18	11	12	18	—
			C.U.	—	—	—	—	8.7	16.7	—

— : not measured.

The content of ^{137}Cs in feces was always below the detectable limit.

Table 7. Weight, and Content of Calcium and Potassium in Diet
(g/day)

Date \ Weight	Wet Weight	Dry Weight	Ash Weight	Calcium	Potassium
1962. May 21	1881	460	15.3	—	—
22	2082	420	15.7	—	—
23	1690	465	18.1	—	—
24	2350	625	23.2	—	—
25	2083	541	15.8	—	—
average	2017	503	17.6	0.64	1.40
1962. May 28	2048	493	17.7	—	—
29	2194	537	27.0	—	—
30	1937	491	9.3	—	—
31	2009	428	19.9	—	—
June 1	1935	388	14.3	—	—
average	2105	467	17.6	0.52	—
1962. Aug. 27	2309	554	26.3	—	—
28	2182	439	17.9	—	—
29	2066	466	15.2	—	—
30	1984	363	20.4	—	—
31	2048	469	20.1	—	—
average	2118	458	20.0	0.48	1.64
1962. Sep. 17	2017	411	20.0	—	—
18	2525	547	24.5	—	—
19	2339	584	18.5	—	—
20	2032	421	18.6	—	—
21	2068	462	21.0	—	—
average	2196	485	20.6	0.66	1.64
1962. Oct. 8	1778	427	17.9	—	—
9	2035	514	18.5	—	—
10	2034	451	16.2	—	—
11	2101	504	24.2	—	—
12	1988	440	16.3	—	—
average	1989	467	18.6	0.52	1.52
1962. Nov. 12	1716	369	16.4	—	—
13	2487	537	17.5	—	—
14	1919	439	20.3	—	—
15	2035	538	17.3	—	—
16	2103	477	15.3	—	—
average	2052	472	17.3	0.60	1.62
1963. Feb. 4	2620	527	20.9	—	—
5	2776	608	18.7	—	—
6	2547	458	20.1	—	—
7	2608	548	21.5	—	—
8	3212	506	18.0	—	—
average	2755	529	19.8	0.53	2.58
Average	2180	483	18.8	0.56	1.73

— : not measured.

Table 8. Volume of Urine, Weight of Phosphate Precipitate, and Contents of Calcium and Potassium in Urine (ml or g/day/person)

Subject / Date	I				II				III				IV				V			
	Volume	Ppt.	Ca	K	Volume	Ppt.	Ca	K	Volume	Ppt.	Ca	K	Volume	Ppt.	Ca	K	Volume	Ppt.	Ca	K
1962. May 21	1310	—	—	—	810	—	—	—	1050	—	—	—	600	—	—	—	750	—	—	—
22	790	—	—	—	1030	—	—	—	1060	—	—	—	1860	—	—	—	870	—	—	—
23	1530	—	—	—	1060	—	—	—	1490	—	—	—	1030	—	—	—	660	—	—	—
24	1450	—	—	—	1020	—	—	—	1130	—	—	—	1370	—	—	—	1220	—	—	—
25	1440	—	—	—	1360	—	—	—	1300	—	—	—	1480	—	—	—	760	—	—	—
average	1310	2.4	—	—	1060	1.6	—	—	1210	1.2	—	—	1270	1.5	—	—	850	1.7	—	—
1962. May 28	1040	0.9	—	—	1300	0.8	—	—	1050	0.5	—	—	—	—	—	—	1420	0.7	—	—
29	920	0.9	—	—	2440	1.1	—	—	1980	0.9	—	—	—	—	—	—	980	0.6	—	—
30	1230	1.8	—	—	790	0.9	—	—	1260	1.1	—	—	—	—	—	—	570	1.1	—	—
31	610	0.7	—	—	740	0.8	—	—	960	0.6	—	—	—	—	—	—	800	1.1	—	—
June 1	1020	1.2	—	—	1370	1.2	—	—	850	1.0	—	—	—	—	—	—	680	0.8	—	—
average	960	1.1	—	—	1330	1.0	—	—	1220	0.8	—	—	—	—	—	—	890	0.9	—	—
1962. Aug. 27	560	—	—	—	510	—	—	—	1680	—	—	—	820	—	—	—	960	—	—	—
28	650	—	—	—	1060	—	—	—	1180	—	—	—	650	—	—	—	640	—	—	—
29	600	—	—	—	980	—	—	—	870	—	—	—	870	—	—	—	510	—	—	—
30	720	—	—	—	850	—	—	—	860	—	—	—	610	—	—	—	490	—	—	—
31	1020	—	—	—	1460	—	—	—	1080	—	—	—	860	—	—	—	460	—	—	—
average	710	1.0	—	—	970	1.6	—	—	1130	1.1	—	—	760	1.3	—	—	610	1.2	—	—
1962. Sep. 17	900	1.6	—	—	510	1.0	—	—	830	1.5	—	—	—	—	—	—	820	0.8	—	—
18	800	1.2	—	—	880	0.2	—	—	1230	1.4	—	—	—	—	—	—	1060	1.0	—	—
19	910	1.5	—	—	1230	1.5	—	—	1430	1.1	—	—	—	—	—	—	960	1.7	—	—
20	960	1.2	—	—	540	1.5	—	—	1060	0.8	—	—	—	—	—	—	1340	1.4	—	—
21	840	1.2	—	—	1220	1.6	—	—	1140	1.4	—	—	—	—	—	—	610	1.1	—	—
average	880	1.3	—	—	880	1.2	—	—	1140	1.2	—	—	—	—	—	—	960	1.2	—	—
1962. Oct. 8	1300	2.3	0.50	2.2	1130	1.1	0.30	1.2	1080	1.1	0.24	1.0	1140	1.0	0.23	1.1	1000	1.1	0.19	1.1
9	1460	1.0	0.16	1.7	940	1.1	0.20	0.7	1480	0.9	0.18	1.2	1230	0.9	0.24	1.2	970	1.0	0.17	1.1
10	1400	1.2	0.22	1.9	1520	2.7	0.28	2.0	1660	1.1	0.24	1.9	1170	1.0	0.24	1.5	940	1.0	0.15	1.2
11	810	1.2	0.15	1.9	2080	0.7	0.28	1.9	1370	1.2	0.26	1.6	1890	1.4	0.27	2.4	700	2.3	0.23	1.2
12	1470	1.3	0.29	2.5	1660	1.7	0.36	0.8	1500	0.9	0.25	1.8	1040	1.6	0.25	1.7	1360	1.5	0.30	2.3
average	1290	1.4	0.26	2.1	1420	1.5	0.28	1.3	1420	1.0	0.23	1.5	1290	1.2	0.25	1.6	990	1.4	0.21	1.4
1962. Nov. 12	760	1.5	0.22	0.9	1870	1.1	0.23	1.8	—	—	—	—	2090	1.1	0.24	1.9	1340	1.1	0.15	1.3
13	1050	1.3	0.19	1.6	1660	1.0	0.22	1.4	—	—	—	—	1700	1.1	0.24	1.5	960	0.1	0.08	0.2
14	1160	1.1	0.20	2.9	1660	1.4	0.29	1.7	—	—	—	—	960	1.0	0.22	1.1	2320	1.5	0.29	1.2
15	950	1.8	0.24	1.4	1770	1.1	0.25	1.5	—	—	—	—	1150	1.4	0.27	1.3	1330	1.1	0.22	0.8
16	1140	1.4	0.19	2.4	1780	1.2	0.21	—	—	—	—	—	1670	1.1	0.24	1.4	950	1.1	0.13	1.8
average	1010	1.4	0.21	1.8	1750	1.2	0.24	1.3	—	—	—	—	1510	1.1	0.24	1.4	1380	1.0	0.17	1.1
1963. Feb. 4	1180	2.1	0.30	2.2	1220	1.0	0.23	1.3	1240	0.8	0.23	1.1	1220	1.4	0.26	1.5	—	—	—	—
5	1320	1.4	0.17	1.5	2420	1.1	0.23	1.8	2140	0.8	0.20	1.6	1440	0.9	0.17	2.0	—	—	—	—
6	1440	1.6	0.19	3.2	1260	0.9	0.13	1.6	1440	0.4	0.13	1.3	1460	1.1	0.20	1.8	—	—	—	—
7	1110	1.4	0.17	1.4	1840	1.7	0.22	2.3	1630	1.4	0.20	1.7	1610	1.2	0.21	1.9	—	—	—	—
8	1300	2.3	0.30	2.2	1640	1.1	0.17	1.8	1580	1.0	0.18	1.7	1580	1.3	0.21	2.3	—	—	—	—
average	1270	1.8	0.23	2.1	1080	1.2	0.20	1.8	1610	0.9	0.19	1.5	1460	1.2	0.21	1.9	—	—	—	—
Average	1060	1.5	0.23	2.0	1300	1.3	0.24	1.5	1290	1.0	0.21	1.5	1260	1.3	0.23	1.6	950	1.2	0.19	1.2

— : not measured.

Table 9. Weight of Ashes, and Contents of Calcium and Potassium in Feces
(g/day)

Subject / Date	I			II			III			IV			V		
	Feces Ash	Ca	K	Feces Ash	Ca	K	Feces Ash	Ca	K	Feces Ash	Ca	K	Feces Ash	Ca	K
1962. May 21	4.1	—	—	1.8	—	—	3.5	—	—	3.2	—	—	4.9	—	—
22	3.4	—	—	3.5	—	—	2.8	—	—	1.0	—	—	2.5	—	—
23	2.9	—	—	1.5	—	—	2.7	—	—	0.8	—	—	3.8	—	—
24	3.4	—	—	2.6	—	—	2.1	—	—	1.2	—	—	4.8	—	—
25	3.3	—	—	6.3	—	—	2.5	—	—	1.5	—	—	4.5	—	—
average	3.4	0.39	—	3.1	0.43	—	2.7	0.25	—	1.5	0.23	—	4.1	0.28	—
1962. May 28	3.0	—	—	6.7	—	—	2.5	—	—	—	—	—	3.7	—	—
29	4.2	—	—	3.5	—	—	3.7	—	—	—	—	—	1.5	—	—
30	3.7	—	—	2.4	—	—	4.0	—	—	—	—	—	3.6	—	—
31	3.9	—	—	5.3	—	—	2.6	—	—	—	—	—	0.5	—	—
June 1	3.4	—	—	4.1	—	—	2.8	—	—	—	—	—	6.9	—	—
average	3.6	0.17	0.56	4.4	0.21	0.40	3.1	0.23	0.43	—	—	—	3.2	0.20	0.48
1962. Aug. 27	0.0*	—	—	0.0*	—	—	2.7	—	—	1.6	—	—	3.2	—	—
28	2.4	—	—	6.9	—	—	3.2	—	—	4.3	—	—	0.0*	—	—
29	2.7	—	—	0.8	—	—	2.7	—	—	3.9	—	—	4.9	—	—
30	1.6	—	—	2.0	—	—	2.1	—	—	0.0*	—	—	6.2	—	—
31	3.2	—	—	3.6	—	—	4.2	—	—	0.0*	—	—	0.0	—	—
average	2.0	0.22	0.28	2.7	0.35	0.24	3.0	0.18	0.44	2.0	0.28	0.24	2.9	0.15	0.43
1962. Sep. 17	2.4	—	—	0.0*	—	—	1.6	—	—	—	—	—	3.6	—	—
18	2.8	—	—	6.1	—	—	2.9	—	—	—	—	—	0.0*	—	—
19	2.3	—	—	3.7	—	—	2.4	—	—	—	—	—	2.5	—	—
20	3.6	—	—	3.9	—	—	2.6	—	—	—	—	—	0.0*	—	—
21	2.5	—	—	1.7	—	—	2.0	—	—	—	—	—	6.8	—	—
average	2.7	0.19	0.33	3.1	0.29	0.23	2.3	0.26	0.35	—	—	—	2.6	0.29	0.33
1962. Oct. 8	3.5	—	—	4.0	—	—	2.3	—	—	0.0*	—	—	0.0*	—	—
9	2.9	—	—	0.0*	—	—	2.3	—	—	1.7	—	—	4.2	—	—
10	0.9	—	—	3.5	—	—	3.9	—	—	0.0*	—	—	5.0	—	—
11	3.6	—	—	2.8	—	—	3.6	—	—	3.1	—	—	1.0	—	—
12	5.3	—	—	4.3	—	—	3.4	—	—	6.8	—	—	0.0*	—	—
average	3.2	0.29	0.50	2.9	0.43	0.27	3.1	0.19	0.48	2.2	0.28	0.23	2.0	0.14	0.31
1962. Nov. 12	3.4	—	—	3.2	—	—	—	—	—	1.7	—	—	0.0*	—	—
13	5.2	—	—	3.0	—	—	—	—	—	0.0*	—	—	4.1	—	—
14	4.3	—	—	4.2	—	—	—	—	—	12.7	—	—	4.6	—	—
15	3.8	—	—	4.0	—	—	—	—	—	3.2	—	—	4.4	—	—
16	4.0	—	—	2.9	—	—	—	—	—	0.0*	—	—	0.0*	—	—
average	4.1	0.30	0.68	3.5	0.42	0.43	—	—	—	3.5	0.49	0.40	2.6	0.19	0.37
1963. Feb. 4	4.0	—	—	4.9	—	—	6.2	—	—	0.0*	—	—	—	—	—
5	4.8	—	—	0.0*	—	—	3.2	—	—	6.5	—	—	—	—	—
6	3.0	—	—	6.2	—	—	3.3	—	—	0.0*	—	—	—	—	—
7	4.6	—	—	5.3	—	—	5.8	—	—	7.8	—	—	—	—	—
8	3.5	—	—	1.2	—	—	1.0	—	—	2.2	—	—	—	—	—
average	4.0	0.24	0.79	3.5	0.54	0.44	3.9	0.30	0.34	3.3	0.45	0.66	—	—	—
Average	3.3	0.26	0.52	3.3	0.38	0.34	2.7	0.23	0.41	2.5	0.35	0.38	2.9	0.21	0.38

— : not measured.
* no fecal excretion.