Observations of the Fall-out Radionuclides at Kanazawa

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Observations of the fall-out radionuclides at Kanazawa

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From January 1963 to May 1965, four events in regard to the radioactive fallout were experienced at Kanazawa owing to the nuclear test explosions in foreign countries. In these cases, some studies were carried out by the members of the radiochemical laboratory of Kanazawa University and their results are reported here.

1. Radionuclide fractionation seen in the fallout accompanied with the heavy snow fall in January, 1963.

In the middle of January 1963, there was the heavy snow fall at the Hokuriku Districts of Japan, including our city, Kanazawa. Several radionuclides were observed by means of gamma-ray spectrometer in the samples brought with snowfall and unusual environmental radiation dose of 0.016 mr/h was measured at this area by survey meter on February 1. These radionuclides might be originated from the Russian large scale nuclear test air-burst in the autumn of 1962. The relative contents of some radionuclides in various samples were studied and the fractionation effect for radionuclides produced by nuclear explosion was observed in those samples. This fractionation process may be caused through a whole stage of their history, that is, from their production to their fall with snow. Furthermore, when snow thaws and that water passes into the soil of surface area, the secondary fractionation may occur owing to the different properties of these radionuclides in solution. These informations for each radionuclide in various samples are useful to deal with the health physical problems related to radiological hazards.

Gamma scintillation counter of Nuclear Corp. with 1 $''\phi \times 1''$ NaI crystal was used to measure the gross activity and 100 channel pulse height analyzer of Ten AN-100 type (Kobe Kogyo Corp.) with $1^3/4\ ''\phi \times 2''$ NaI well type crystal was also used for gamma spectrometric measurement of various samples.

All samples studied were collected at the old campus of the Faculty of Science in Sengoku-machi of Kanazawa city. The samples (a) and (b), whose spectrums were shown in Fig. 1, were the snow samples deposited on the same place at the playground, but the former was sampled from the surface of deposited snow and the latter from the layer at 50 cm depth. Both samples were melted with heating, respectively

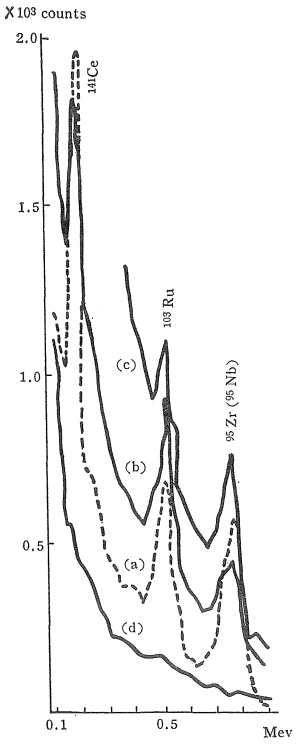

Fig. 1. Gamma ray spectra of snow and soil

(a) the surface snow sample (12′50″)
(b) the 50cm depth snow sample (46′31″)
(c) the soil sample (60′)
(d) the drinking water sample (20′)
( ) ; measuring time

and one litre of each sample was evaporated to dryness in order to measure its gamma radioactivity. The sample (a) had 2.6 × 10^3 counts per minute radioactivity and the sample (b) showed 0.94 × 10^3 cpm (background count : 0.57 × 10^3 cpm). From the gamma ray spectrums shown in Fig. 1, it was found that Ce-141 (its half life: 33.1 days), Ru-103 (39.8 d) and Zr-95 - Nb-95 pair (their half lives: 65 d and 35 d respectively) were predominant components, but the relative activities of these nuclides were different from each other. Ratios of peak heights due to these nuclides are estimated graphically as follows. The Ru-103 : Zr-95 : Ce-141 ratios are found to be 1 : 1.1 : 2.3 in the sample (a) and 1 : 0.5 : 1.1 in the sample (b). For comparison, gamma ray spectrums of drinking waters (one litre) in Kanazawa city and soils collected at ground surface are shown also in Fig. 1. It should be noticed that the peak height due to Zr-95 is more conspicuous than that due to Ru-103 in the soil sample.

To examine the fractionation effects of these radionuclides in various materials, the column bed was prepared with sands of different particle size and cation and anion ion exchangers, and the thawed snow waters was passed into that column bed continuously from the tiled roof of the wooden laboratory building through the polyethylene tube. As shown in the left part of Fig. 2, the column bed was made by the following way. An acryl resin column (4 cm in diameter and 23cm long) was filled successively from the upper part to the lower part with coarse sands of about 30-60 mesh particle size, fine sands of under 60 mesh, cation ion exchanger (Amberlite IR-120 of 30-40 mesh) and finally anion ion exchanger (Dowex 1 × 8 of 200-400 mesh). The upper and lower ends of the column were sealed with nylon of fine net structure and the same net was used at the boundary between different bed fractions. After passing about 100 litres of thawed snow water into the column, the column bed was sliced to every one cm thick layer. Then, the measurement of gross gamma activities and gamma ray spectrometry were carried out for each sliced layer. The results are shown in Fig. 2 and Fig. 3.

Along with these experiments, one litre of the original thawed snow water and the effluent solution from the column (filtrate) were evaporated to dryness and the radioactivity of each sample was measured respectively. Besides these samples, the

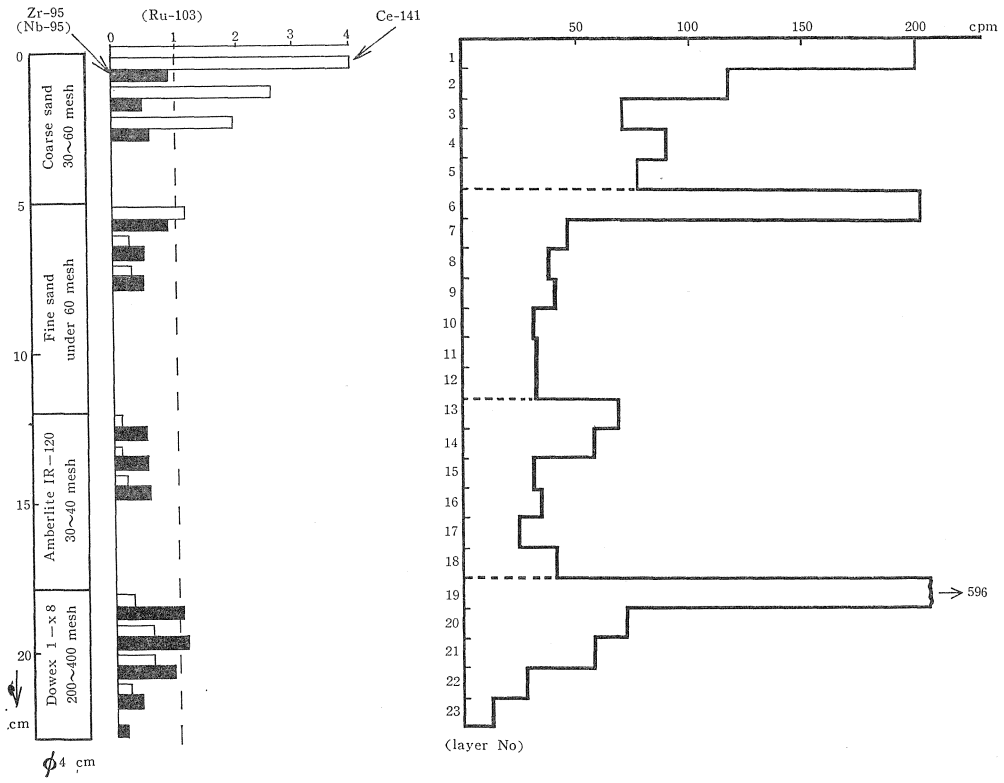

Fig. 2. Column beds and the radioactivity found in each slice of the experiment.

black dust fragment floating on thawed snow water (dust F) was sampled and measured in similar way. These results are shown also in Fig. 3. It is noteworthy that the relative peak heights due to Zr-95 and Ce-141 to that of Ru-103 are much higher in the dust sample (F) having the net gamma activity of 85 cpm as compared with all other samples examined.

From the gamma radioactivity distribution shown in Fig. 2, it is clearly found that the upper layers of each four different bed fraction have relatively high concentrations of radionuclides compared to the lower layers of the same fraction. In the analysis of these results, the following observation should be remarked. After the experiment, the lowest fraction of the column bed, Dowex 1, had been covered with gray fine powders from its upper to middle layer. Such condition might be caused by the migration of fine sands of the upper fraction of the column bed to the anion exchanger fraction through the coarse cation exchanger particles. So, the contribution of fine sands to the upper layer of the anion exchanger fraction must be considered.

Radionuclides observed in the gamma ray spectrums of all samples are mainly the following nuclides: Zr-95 (gamma energy, 0.726 and 0.760 MeV) and Nb-95 (0.768

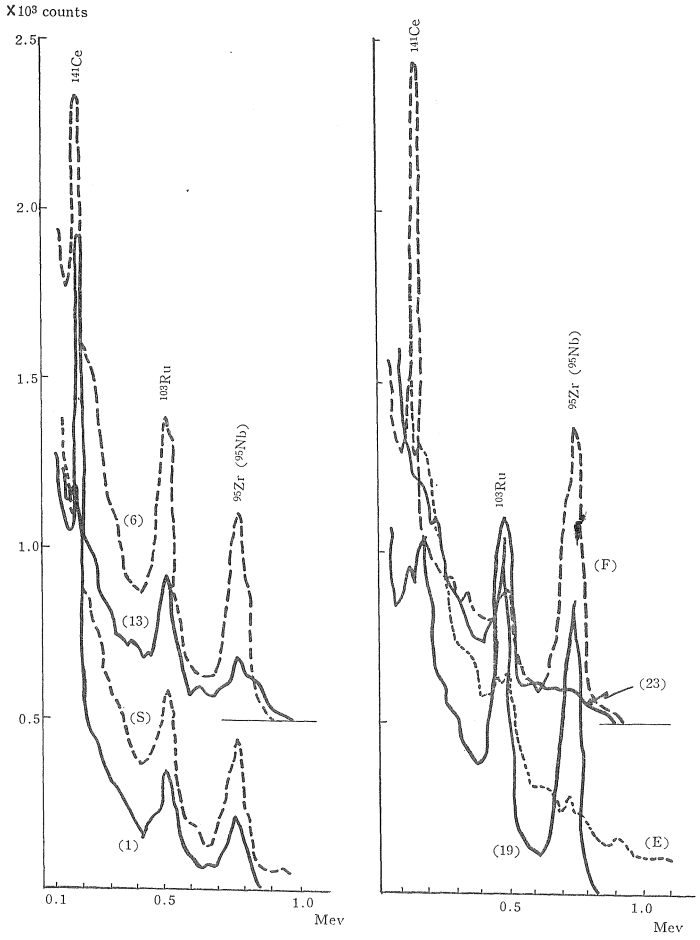

Fig. 3. Gamma ray spectra of each slice

(1)	No. 1 slice (4′1″)	(19)	No.19 slice (2′28″)
(6)	No. 6 slice (8′5″)	(23)	No.23 slice (20′)
(13)	No.13 slice (7′30″)	(F)	the dust sample (3′40″)
(S)	the snow water (20′)	(E)	the filtrate (40′)

( ) ; measuring time

MeV), Ru-103 (0.498 MeV) and Ce-141 (0.145 MeV). The relative activities of these nuclides in each sample are estimated by comparing the peak heights of three peaks corresponding to Zr-95 (Nb-95), Ru-103 and Ce-141 activities in gamma spectrums. Having many different chemical forms in solution, Ru-103 was retained in all samples at different degrees and observed even in the filtrate sample. The presence of Ru-103 in the low radioactive filtrate may be derived from the deficiency of whole exchange capacity of Dowex 1 x 8 resin used or the non-ionic state of this nuclide. Activities due to Zr-95 (Nb-95) was not found in the fitrate, but enriched in the fine sands fraction including the contaminated portion to anion exchanger. Especially, Zr-95 (Nb-95) was much enriched in the dust sample (F). It is noticeable that the activity of

Ce-141 is retained obviously in the upper part of the coarse sands fraction and also in the dust sample (F).

From the results mentioned above, the fractionation of radionuclides in different materials is fairly well recognized. Therefore, if considerable masses of snows have thawed and resulting waters pass into ground, the fractionation of the radionuclides will occur and some nuclides will be captured by soil components. As one example of this case, it is interesting to observe the migration process of radionuclide from snows to ground at the playground where remarkable amounts of snows were dumped. When the thawing of dumped snow had come, two soil samples of ground were collected from the different sites of this area on May 1. These two samples were cut into every one cm fraction from the surface to the deeper part. By the radioactivity measurement of each fraction, it was found that the surface fraction was more radioactive than the other fraction, and Zr-95 (Nb-95) and Ce-141 were clearly enriched compared to Ru-103. Thus, the more effective fractionation had occurred in the soil samples than in the snow samples.

The environmental radiation dose showed somewhat high values around the playground even in July, 1963. The three or four holds higher radioactivities than that of background were measured by survey meter at the places where appreciable dusts accumulated after the thawing of dumped snows.

2. Radionuclides in the fallout from the first Chinese nuclear test explosion

Resulting from the first Chinese nuclear explosion carried out on October 16, 1964, the highly radioactive fallout fell on Kanazawa city two days after this explosion. In order to examine the radionuclides in this fallout, the sample was collected on the roof of the building belonging to the Faculty of Science by sweeping that floor at about 10 o'clock a. m. on October 20. The radioactivity measurement of this sample by well type scintillation counter showed about 75×10^3 cpm as for 5g of this sample on that day. The gamma ray spectra of this sample were recorded by 100 channel PHA at various linear amplifier condition. In each case, the channel-energy calibration curve was made by using several radionuclides (Ce-144, Pa-233, Cs-137, Zn-65 and Co-60) as the standard gamma energy sources. The spectra of the fallout sample at various gamma energy region are showed in Fig. 4 (a), (b), (c), (d). The same sample had the gamma activity of 38×10^3 cpm on October 23 and its spectrum is shown in Fig. 4 (a) as the dotted line for comparison with the spectrum on October 20. From these spectra, the existences of the following radionuclides are confirmed: Np-239 (2.35 d.), Ba-140 and La-140 pair (12.8 d. and 40.2 h. respectively), and I-132 (2.26 h., the daughter of Te-132 having half life of 77.7 h.). The existence of Ce-141 (33.1 d.) having the gamma energy of 0.14 MeV is also revealed by comparing the dotted line spectrum with the former one.

In Fig. 5, another gamma ray spectrum of the rain-out samples are shown. These

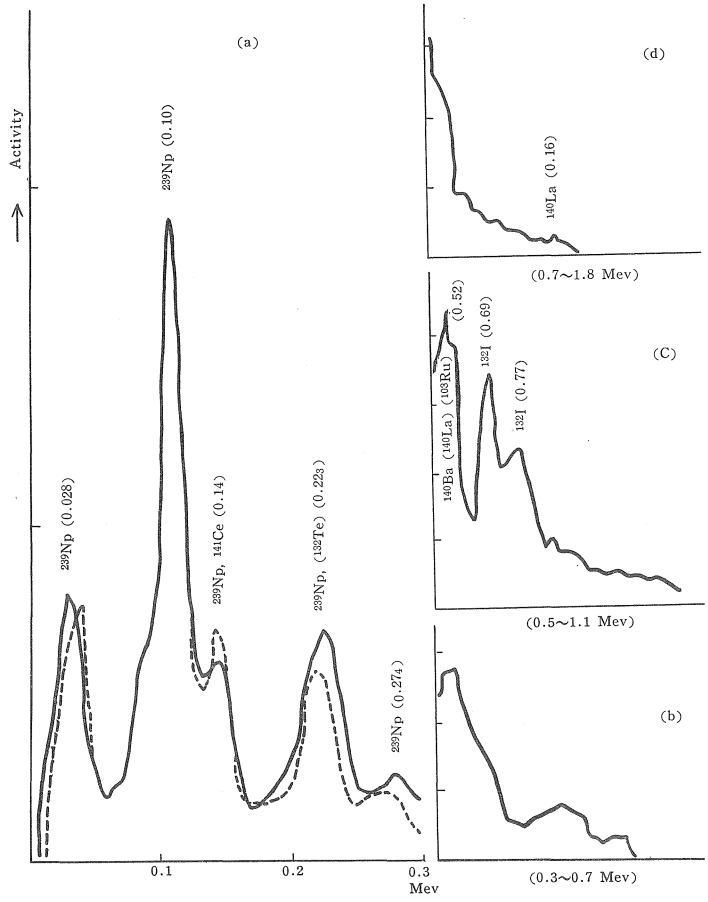

Fig. 4. Gamma ray spectra of the dry fallout sample in various energy regions

samples were collected on the roof of the Ishikawa hygienic laboratory in the different period of October, 1964. The sample (a) is the total rain-out from lst to 16th of this month and the sample (b) from 17th to 29th. Both samples were made by evaporating one litre of the original sample, respectively and subjected to the gamma ray measurement. The gross gamma activities were 750 cpm (B.G. 530 cpm) for the sample (a) and 26 $\times$ 10^3 cpm for the sample (b). From these data and the gamma ray spectra of these samples, it is obvious that the first Chinese nuclear test explosion has a great effect on the radioactivity of the rain-out samples. As these spectra were recorded on October 30, the peaks due to Np-239 which had been seen overwhelmingly in the spectra of October 20, diminished owing to its fairly short half life and the peaks due to Ce-141, La-140 and Zr-95 (Nb-95) were identified in this spectrum.

Besides the gamma ray spectrometric study, the radiochemical analysis was carried out as for the above mentioned sample by the following procedure in order to examine mainly the existence and abundance of various uranium isotopes. The sample was

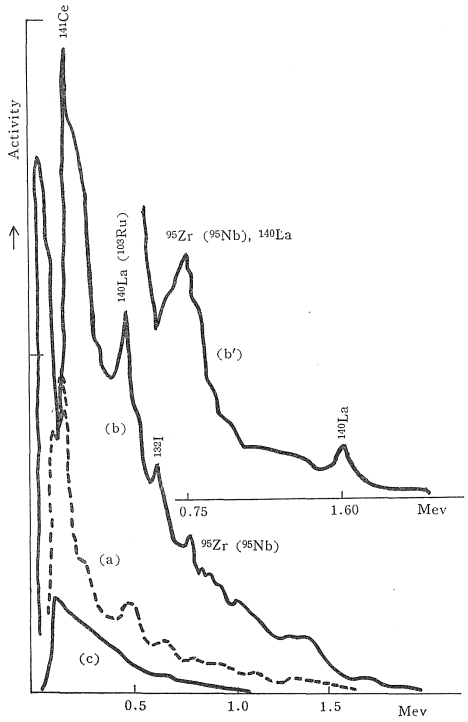

Fig. 5. Gamma ray spectra of the rain-out samples
(a) the sample from lst to 16th, October, 1964
(b) the sample from 17th to 29th, October, 1964; 2^{12} counts full scale recording
(b') the same sample; 2^{10} counts full scale recording
(c) the back ground radioactivity
(All spectra were recorded after the radioactivity measurements for 20 minutes.)

decomposed by aqua regia and evaporated up to nearly dryness. Then, this residue was taken up by concentrated nitric acid and the solution was separated from the residue by filtration. The chemical separation procedure with ethyl acetate solvent extraction and anion exchange was applied to this solution and the main scheme of this procedure is shown in Fig. 6. The final solution leached from Dowex 1 anion exchanger by 0.1 N HCl was converted to the electrolytic solution which contained 0.25 M formic acid and had the definite pH value suitable for effective electrodeposition of uranium. The electrodeposition was carried out on the stainless steel plate (3.5 cm diameter) as cathode for two hours by electric current of 400 mA. The counting source obtained by

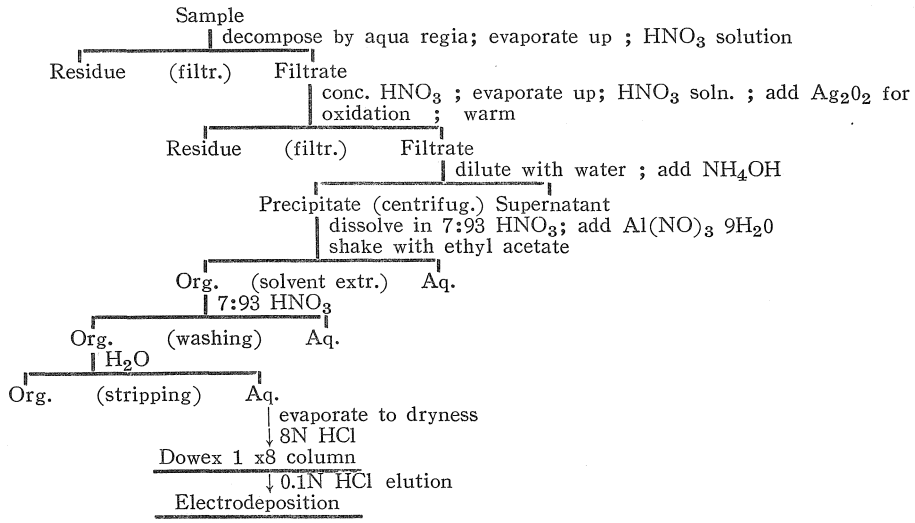

Fig. 6. The chemical separation scheme

this procedure was subjected to the beta and alpha radioactivity measurements. The beta activity was 115.4 cpm at the time immediately after the above mentioned procedure and decayed exponentially by the half life nearly equal to that of U-237 as shown in Fig. 7. So, the existence of U-237 (6.7 d.) is unquestionable and it should be remarked that the nuclear reaction, U-238 (n, 2n) U-237, by fast neutron had occured in the first Chinese test explosion together with the reaction, U-238 (n, γ) U-239→Np-239, by thermal neutron.

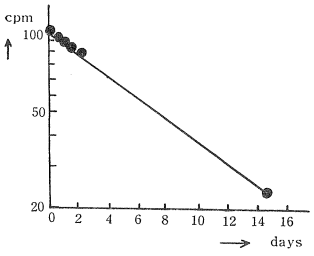

Fig. 7. Uranium-237 decay curve

To study the alpha emitters in the electrodeposited source, the alpha pulse spec-

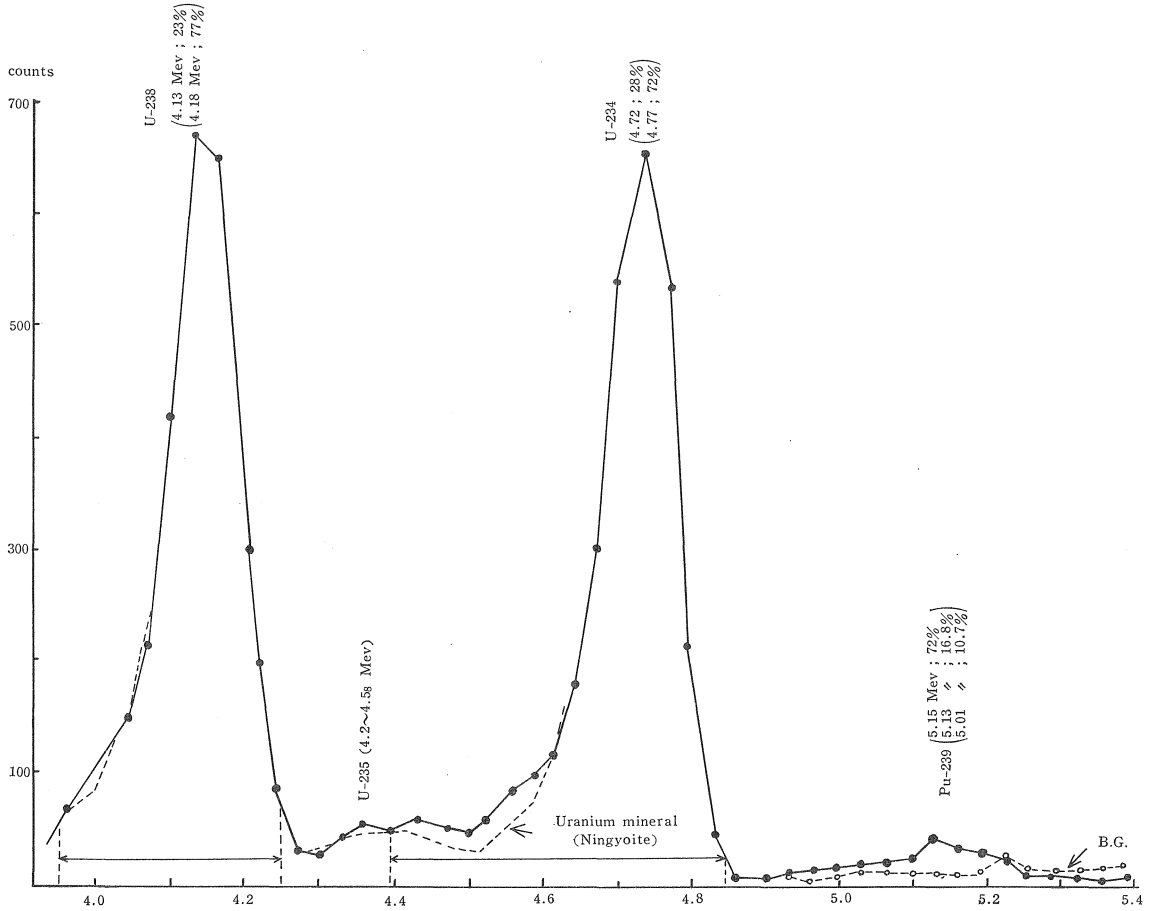

Fig. 8. Alpha ray spectrum

trometry was carried out by the combination of Frish grid ionization chamber (Osaka Denpa Corp.) with the rise time converter and 100 channel PHA. From the alpha spectrum shown in Fig. 8., it is found that the ratio of U-234 to U-238 is nearly unity in activity unit ratio and therefore the natural uranium had been contained in fairly large amount in the first Chinese nuclear bomb. This fact was proofed also by the existence of conspicuous gamma energy peaks due to Np-239 in the early gamma ray spectrum of the same sample. The small peak in the about 5.1 Mev region of the alpha spectrum is considered to be attributable to Pu-239, but it could not be confirmed whether this nuclide produced as the decay product of Np-239 found in the fallout or the original nuclear bomb had contained Pu-239 in some quantity before that explosion.

3. Radionuclides in the fallout from the underground nuclear test explosion in U.S.S.R..

The rain-out sample collected at Kanazawa in the middle of January, 1965, showed the radioactivity due to the fission products probably originated from the underground nuclear test explosion which was reported to be carried out in U.S.S.R. some days ago. The gamma spectrum of this sample recorded on January 22, showed obviously the existences of Te-132 (I-132) and Ba-140 (La-140) as shown in Fig. 9. Furthermore, it was confirmed from the spectrum recorded on February 23 that this same sample had contained such radionuclides as Ce-141, Ru-103 and Zr-95 (Nb-95) as shown in

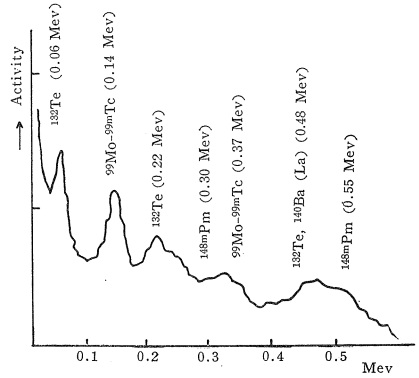

Fig. 9. Gamma ray spectrum of the fallout sample recorded on Jan. 22, 1965.

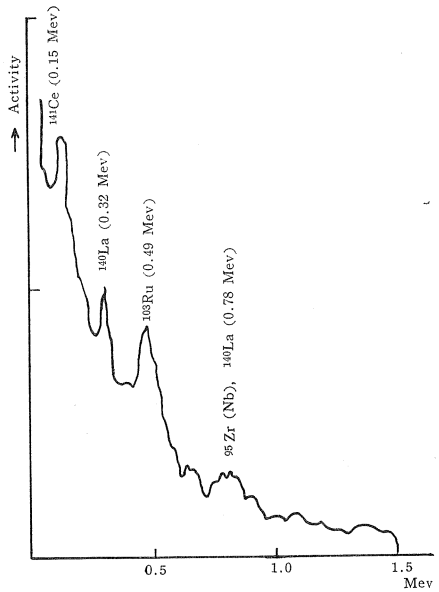

Fig. 10. Gamma ray spectrum of the fallout sample recorded on Feb. 23, 1965.

Fig. 10. As contrast with the first Chinese nuclear test explosion, it was noticeable that the gamma activity peak of Np-239 had not come out clear in the spectrum of the energy region up to 500 keV which was recorded few days after the nuclear explosion and shown in Fig. 9.

4. The radioactive fallout from the second Chinese nuclear test explosion

Fig. 11. Gamma ray spectra of the fallout samples
(a) the sample collected on May 17; measured on that day for 34 minutes
(b) the sample collected on May 22; measured on May 24 for 26 minutes

It was reported that the second Chinese nuclear explosion was carried out on May 14, 1965. The radioactive fallout was collected on the roof of the building belonging to the Faculty of Science of our University on 17th of this month. And the other sample was collected on 22th of this month. The gamma spectrums of these samples were shown in Fig. 11. The absence of the peak due to Np-239 in these spectrum should be remarked in contrast with the spectrum of the fallout from the first Chinese nuclear test explosion. The gross gamma activities of these samples were 1.26×10^3 cpm/4.8gram and 2.3×10^3 cpm/9.5gram respectively on May 24.