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The Analysis of Th in the Korean Total Diet Sample by RNAA

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Abstract

In order to estimate the degree of intake of ^{232}Th through daily diet, a korean total diet sample was collected and made after the investigation of the amount of consumption of daily diet which is dependent on the ages of 20's to 50's. For Th analysis, the RNAA method was applied and NIST SRM 1575, Pine Needle was used as quality control materials. The result of the SRM analysis was compared with a certified value. The relative error was 5%. The determination of the Th in the korean total diet sample was carried out under the same analytical condition and procedure with SRM . As a result of the korean total diet sample, the concentration of Th was in 3.4 ± 0.2 ppb and the amount of daily intake of Th by the diet is found to be 0.67 μg per day. Radioactivity by Th intake was estimated to be about 2.7 mBq per person per day and annual dose equivalent was revealed as 0.73 μSv per person.

1. Introduction

Th-232 is a radioactive element which has about 4075 Bq/g of specific activity and incurs internal doses by inhalation of airborne particulate matter , by intake of daily diet and by industrial activity [1]. Therefore, it is necessary to apply an accurate and precise analytical method for Th and important to assess the radiation doses from this natural source in human body.

For this purpose, RNAA(Radiochemical Neutron Activation Analysis) which has the advantage of high sensitivity and selectivity can be applied as a relevant method [2]. In previous study [3], this possibility showed. Meanwhile, the Korean total diet was prepared, which was based on the food daily data of the ages of 20's to 50's, 77 adults in Korea. The average amount of daily food consumption was 1700g. Table 1 shows the data for the daily food consumption of Korean people in 1997.

In this study, with the determination of the concentration of Th in the Korean total diet by using RNAA, it was intended to estimate the daily intake of Th and assess the radiation dose.

2. Experimental

2.1 Sample Preparation

To prepare the Korean total diet for analysis of Th, the diet sample was purchaesd in markets and pretreated by the following procedures ; for homogenization, diet samples were mixed by blender and freeze dried for 30 hours at -80 ℃. After freeze-drying, the weight of the total diet was reduced from 1120.0 g to 130.2g. The freeze dried sample was finely ground by using an agate motor. The pretreated Korean total diet sample was prepared in a polyethylene capsule, washed by 1N HNO_3 for irradiation. NIST SRM 1575 Pine Needle were done as well after drying for 2 hrs at 80℃ in an oven in accordance with the recommended procedures of NIST certificate.

2.2 Sample Irradiation and Radioactivity Measurement

The prepared diet sample was irradiated under the thermal flux of 1.7 X 10^{13} $n/cm^2 \cdot sec$ for 4 hours using PTS(Pneumatic Transfer System) of the HANARO research reactor. NIST SRM samples were also irradiated in the same condition. These samples were allowed to cool for more than 10 days to reduce interfering activities of medium-lived nuclides(^{24}Na, ^{82}Br etc).

Thorium was determined by using the nuclear reaction of

$$^{232}Th\ (n,\gamma)\ ^{233}Th \xrightarrow{\beta^-} {}^{233}Pa\ (\ T_{1/2} = 27d,\ E_\gamma = 312\ keV)$$

All samples were analyzed by estimating the ^{233}Pa -activity. For the γ-ray measurement of ^{233}Pa and other nuclides, a HP-Ge semiconductor(EG & G ORTEC, GEM25185, 1.85keV resolution at 1332keV of Co-60, Peak to Compton ratio 45 : 1) and 8K MCA(MCB 919, EG & G ORTEC) were used.

2.3 Radiochemical Separation Procedure

The RNAA method was applied for Th analysis [4]. The flow scheme on a separation procedure of Th is presented in Figure 1. Subsequent separation procedures are as follows ; samples were digested in concentrated HNO_3. Add 40 mg of Mn carrier and heat the solution to dryness. Then add 25ml of 4M HNO_3 and gently warm. The MnO_2 precipitaion is then centrifuged and supernatant discarded. The precipitation is dissolved by the addition of a few drops of H_2O_2. To this solution, 7 ml of H_2SO_4 is added and about 100 mg of K_2SO_4. The soultion is gently warmed and 40 mg of Ba carrier is added dropwise and $BaSO_4$ precipitaion appears. The $BaSO_4$ precipitation get redissolved. This solution is cooled and 40 - 50 ml of water is added to it along with constant stirring. The BaSO4 precipitation reappears. This precipitation is filtered through whatmann 542 filterpaper and counted for 312 keV γ-ray peak.

3. Results and Discussion

For the quality control, the concentration of Th in NIST SRM 1575, Pine Needle was determined to check accuracy and precision of the RNAA method. The analytical value was 35 ± 3 ppb. Compared with the certified value (37 ± 8 ppb), relative error was 5%. If the uncertainty would be taken into account, our value was in good agreement with the certified value. Fig 2. shows the gamma-ray spectrum of the Th analysis in SRM Pine Needle with analytical condition.

As a result of the duplicate analysis of the Korean total diet sample, the analytical value was 3.4 ± 0.2 ppb. Fig 3. shows the gamma-ray spectrum of the Th analysis in the Korean Total diet sample. If the average amount of daily food consumption is 1700g, for the calculation of daily Th intake by diet, the Th intake by the Korean total diet was estimated to be 0.67 μg per person per day. Converting this into radioactivity, the value is about 2.7 mBq per person per day. The effective dose equivalent by ^{232}Th intake was known as 7.38 x 10^{-7} Sv/Bq [1]. Consequently, it can be concluded that annual dose equivalent by ^{232}Th in daily diet is about 0.73 μSv per person. Dietary intakes of Th for Japanese have been reported in limited numbers. The mean daily intake and its standard deviation, 0.41 ± p0.21 μg(1.7 ± 0.8 mBq) per person, were reported from a duplicate portion study covering a wide range of Japan [5]. Compared with Japanese value, Our value was estimated to be slightly higher but not significantly different.

From these results, it was clarified that the RNAA method can be applied effectively for the determination of Th in real samples which have ppb or sub-ppb concentrations. In the future, for the assessment of radiation dose from natural source ,U as well as Th, the RNAA will be effectively applied through lots of practical sample analysis.

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Reference

1. Bernard Shleien, Pharm. D. "The health physics and radiological health handbook" , revised edition, Scinta inc., USA, 1992
2. Zeev B. Alfassi, Activation analysis vol I, CRC
3. Proceedings of the Korean Nuclear Society Autumn Meeting, Seoul, Korea, 1998
4. H.S. Dang, D.D. Jaiswal etal, "A sensitive method for the determination of Th in body fluids" Health Physics, Vol 57, No. 3, 393-396, 1989
5. K. SHIRAISHI, Y. IGARASHI, Y. TAKAKU etal, Health Physics, 63 (1992) 187.

Table 1. Average consumption frequency of different food items for one day representative mixed diet of adult Korean and weight of each item to prepare one day Korean representative total diet.

	50's	40's	30's	20's	overall
boiled rice	2.33	2.48	2.14	1.76	2.18 X 210g = 457.8g
kimch'i	2.20	2.64	2.40	1.81	2.26 X 60g = 135.7g
soybean paste stew	0.26	0.68	0.60	0.27	0.45 X 180g = 81g
kimch'i stew	0.36	0.30	0.31	0.23	0.30 X 180g = 54g
fish stew	0.17	0.27	0.17	0.20	0.20 X 180g = 36g
seaweed soupl	0.21	0.07	0.10	0.12	0.13 X 350g = 45.5g
broth (meat juice)	0.24	0.32	0.29	0.31	0.29 X 350g = 101.5g
bean-sprout soup	0.10	0.20	0.13	0.07	0.13 X 350g = 45.5g
bean curd stew	0.02	0.09	0.08	0.03	0.06 X 180g = 10.8g
lamyeon	0.10	0.11	0.14	0.24	0.15 X 120g = 18g
noodles with bean sauce	0.02	0.05	0.04	0.07	0.05 X 520g = 26g
wheat noodles	0.00	0.02	0.00	0.04	0.02 X 750g = 15g
noodles	0.06	0.07	0.17	0.03	0.08 X 90g = 7.2g
Chinese-style hotchpotch	0.00	0.04	0.01	0.02	0.02 X 790g = 15.8g
meat and Chinese noodles	0.14	0.02	0.01	0.05	0.06 X 200g = 12g
pizza	0.02	0.00	0.00	0.05	0.02 X 200g = 4g
hamburger, sandwiches	0.02	0.00	0.01	0.09	0.03 X 200g = 6g
cake	0.06	0.04	0.02	0.01	0.03 X 100g = 3g
pie	0.00	0.02	0.00	0.02	0.01 X 100g = 1g
doughnut	0.04	0.02	0.01	0.01	0.02 X 100g = 2g
table bread	0.00	0.09	0.05	0.09	0.06 X 100g = 6g
cabbage	0.00	0.02	0.01	0.12	0.04 X 70g = 2.8g
radish	0.18	0.09	0.24	0.20	0.18 X 70g = 12.6
Welsh onion	0.04	0.05	0.10	0.05	0.06 X 20g = 1.2g
mushroom	0.06	0.07	0.01	0.09	0.06 X 70g = 4.2g
bean sprouts	0.08	0.09	0.14	0.13	0.11 X 70g = 7.7g
lettuce	0.04	0.07	0.14	0.09	0.09 X 40g = 3.6g
garlic	0.04	0.04	0.04	0.04	0.04 X 10g = 0.4g
red pepper	0.02	0.09	0.08	0.06	0.06 X 20g = 1.2g
cucumber	0.04	0.09	0.06	0.16	0.09 X 70g = 6.3g
sesame leaf	0.12	0.00	0.07	0.06	0.06 X 20g = 1.2g
spinach	0.10	0.21	0.13	0.09	0.13 X 70g = 9.1g
carrot	0.10	0.00	0.01	0.07	0.05 X 70g = 3.5g
onion	0.02	0.00	0.01	0.07	0.03 X 40g = 1.2g
pumpkin	0.02	0.04	0.01	0.02	0.02 X 70g = 1.4g
broad bellflower	0.16	0.00	0.02	0.05	0.06 X 70g = 4.2g

Table 1. (continued).

	50's	40's	30's	20's	overall
bean curd	0.31	0.86	0.46	0.37	0.50 X 80g = 40g
beef	0.43	0.52	0.19	0.11	0.31 X 60g = 18.6g
pork	0.20	0.38	0.44	0.37	0.35 X 60g = 21g
chicken	0.04	0.11	0.19	0.08	0.13 X 60g = 7.8g
* egg	0.30	0.38	0.71	0.49	0.47 X 50g = 23.5g
tuna	0.06	0.00	0.07	0.04	0.04 X 70g = 2.8g
mackerel	0.18	0.30	0.18	0.07	0.18 X 70g = 12.6g
anchovy	0.18	0.27	0.12	0.06	0.16 X 13g = 2.1g
cuttlefish	0.04	0.02	0.06	0.16	0.07 X 70g = 4.9g
walleye pollack	0.08	0.00	0.04	0.08	0.05 X 70g = 3.5g
mackerel pike	0.00	0.04	0.00	0.01	0.01 X 70g = 0.7g
shellfish	0.04	0.07	0.00	0.02	0.03 X 30g = 0.9g
fish ball	0.00	0.02	0.12	0.05	0.05 X 50g = 2.5g
laver	0.71	0.95	0.74	0.42	0.71 X 3g = 2.1g
brown seaweed	0.06	0.20	0.08	0.05	0.10 X 70g = 7g
green layer	0.02	0.04	0.00	0.04	0.03 X 70g = 2.1g
** milk	48.98	140.18	20.24	96.19	76.40
icecream	0.00	0.00	0.04	0.09	0.03 X 100g = 3g
yogurt	0.33	0.34	0.15	0.26	0.27 X 180g = 48.6g
apple	0.71	0.59	0.58	0.23	0.53 X 100g = 53g
persimmon	0.51	0.30	0.10	0.09	0.25 X 100g = 25g
orange	0.06	0.39	0.29	0.20	0.24 X 100g = 24g
pear	0.06	0.05	0.10	0.05	0.07 X 100g = 7g
** beer	3.06	2.68	89.29	82.16	44.30
** distilled spirits	21.43	13.39	63.93	31.49	32.56
** raw rice wine	0.00	0.00	0.00	4.86	1.22
** refined rice wine	6.12	0.00	4.76	0.86	2.94
** coffee	29	50	48	77	51
** coke	6	10.5	16.5	39	18
** soft drink	3	3	3	7.5	4.5
** sweat rice drink	6	3	0	1.5	3
** orange juice	0	10.5	22.5	10.5	10.5
** adlay tea	2	0	0	2	1
** green tea	4	4	8	3	5
** tea	0	0	1	3	1

* Number

** Volume in ml

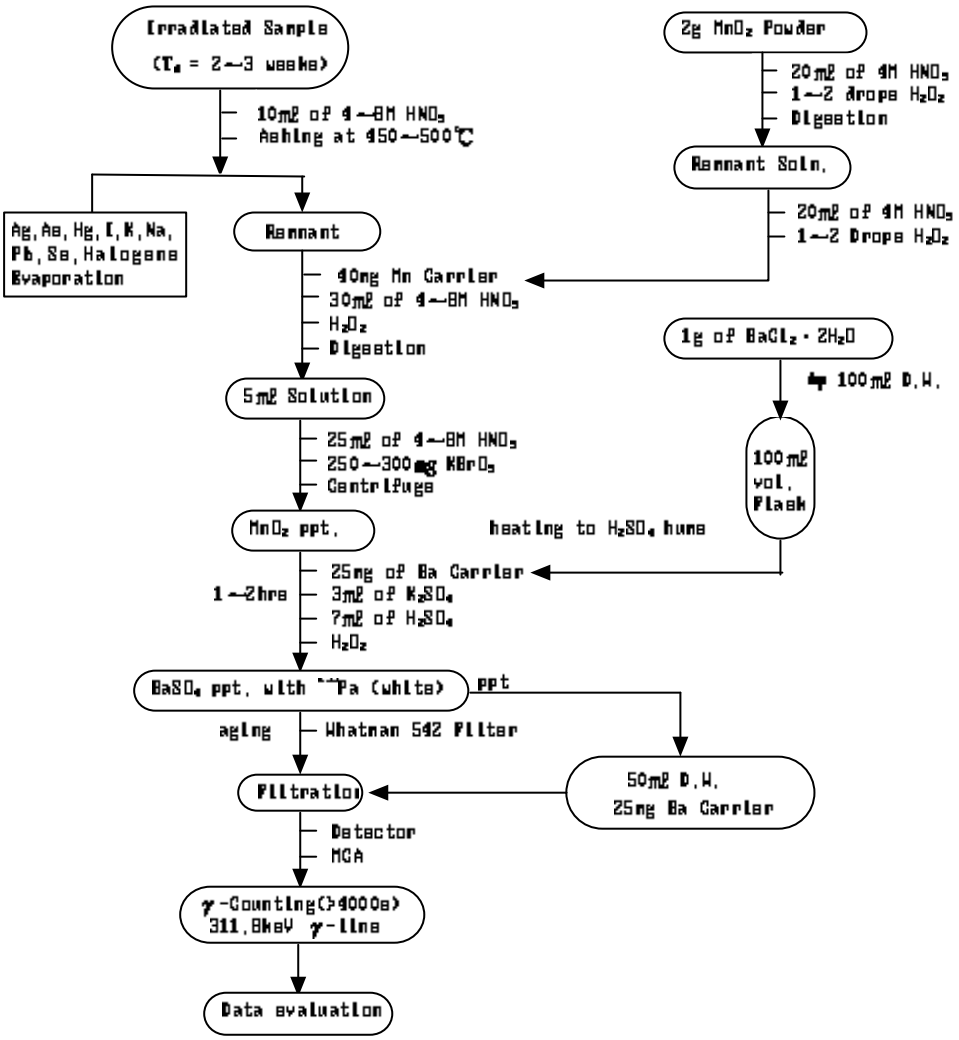

Figure 1. Radiochemical separation scheme for the determination of Th(^{233}Pa)

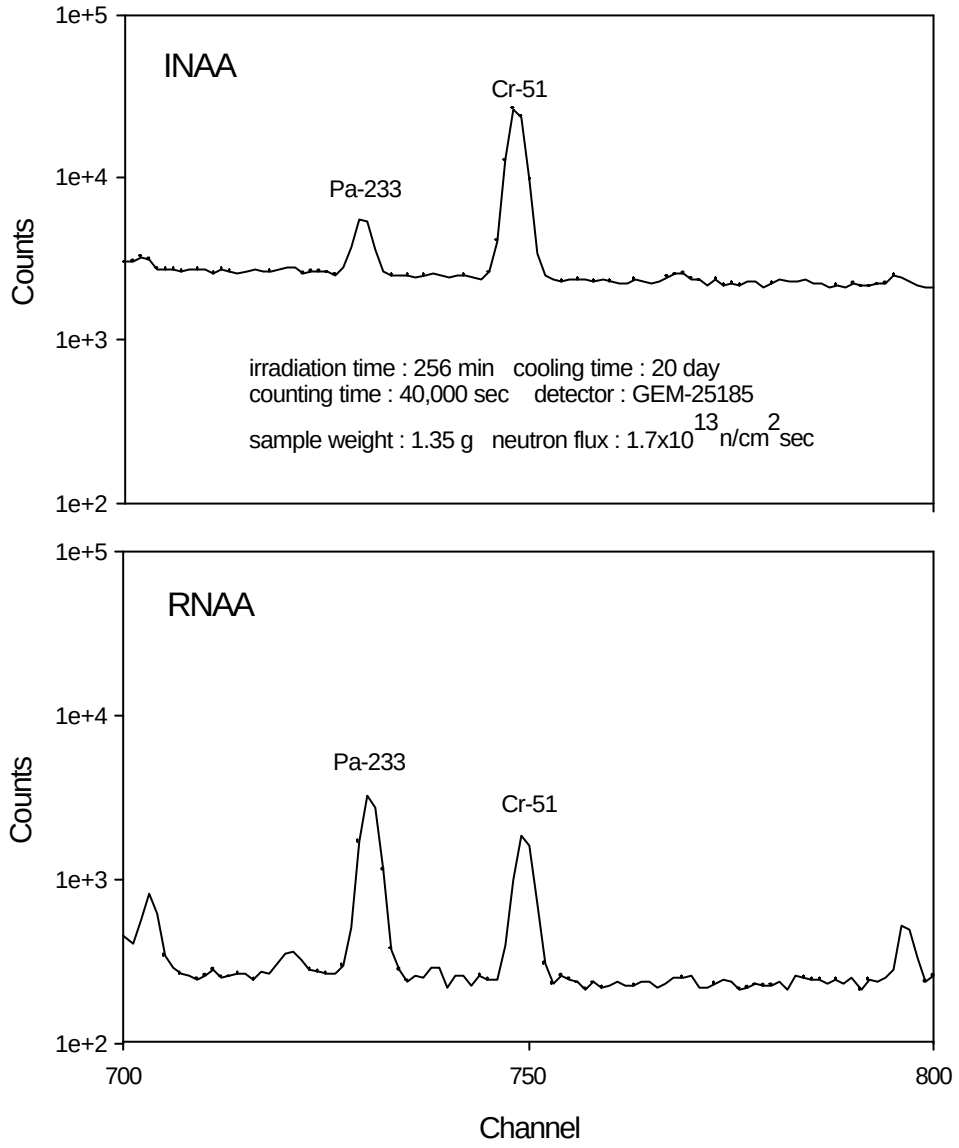

Figure 2. Gamma-ray spectrum of SRM 1575 (pine needles) by INAA and RNAA

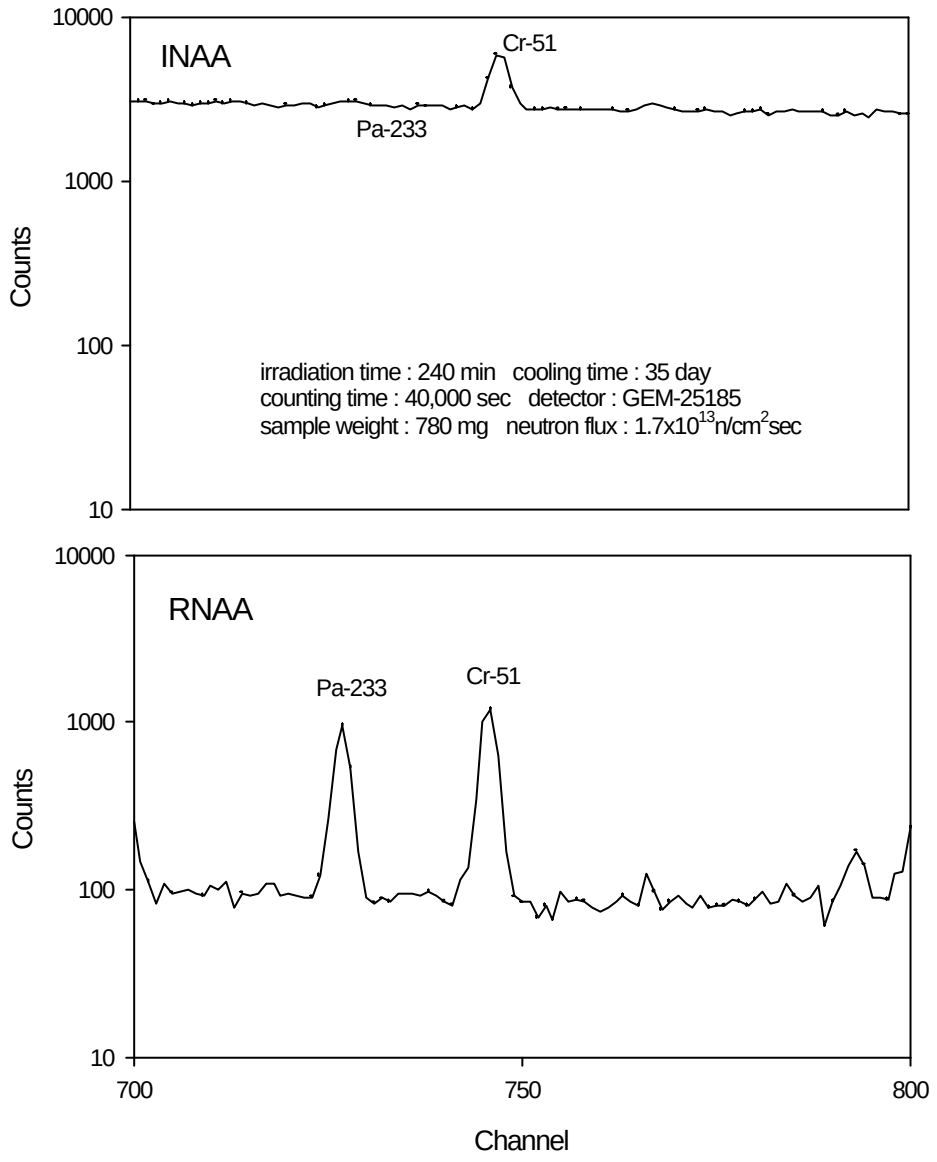

Figure 3. Gamma-ray spectrum of Korean total diet by INAA and RNAA