ORIGINAL

A Study on Natural Absorbed Dose Rate in Air (Using TLDs) and Concentration of Natural Radionuclides in Soil in All Area of the Osaka Pref.

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Absorbed dose rates in air at 40 sites throughout Osaka Pref. were measured by TLDs and a relationship between the dose rates and the rates estimated from concentrations of natural radionuclides in the surface soils collected at the sites was studied. The highest and lowest values of air absorbed dose rates obtained in Osaka Pref. were 114.1 ± 1.0 , 66.3 ± 2.3 nGy/h, respectively, and the former value was obtained at granite zone and the latter at andesite zone. It was found that soil particles smaller than 10 mesh contribute to the dose rates more significantly than those larger than 10 mesh. We found a fairly good correlation between the dose rates and the concentrations of ^{40}K in the surface soils at the sites.

KEY WORDS: absorbed dose rate in air, TLD, Osaka, soil, concentration of natural radionuclides, mineral composition, 10 mesh

I INTRODUCTION

Knowledge on the natural absorbed dose rate

in air is important for an individual and the public from a viewpoint of health physics. An evaluation of practical relationships between the dose rate and the concentration of natural radionuclides in surface soil is useful in estimating easily the dose rate from the concentrations of the nuclides in the surface soil of small amount.

The Osaka Prefecture in October of 1989 had a population of 8,747,000 which is about 7% of the total Japanese population of 123,255,000 and this population is the largest next to the Tokyo metropolitan area. According to field survey data of natural radiation in Japan obtained by S. ABE *et al.*¹⁻³⁾, the dose level in Osaka is about 1.5 times higher than each level in the Tokyo Metropolis, the Kanagawa Pref. and the Chiba Pref. This is due to the fact that the Kansai area including Osaka is composed of granitic zone containing a higher concentrations of natural radionuclides in comparison with other geological

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Fig. 1 A map showing the locations of the monitoring posts.

Fig. 2 A geological map of Osaka Pref.

zones³⁻⁷⁾ while the Kanto area including Tokyo is in volcanic ash zone. The exact determination of the population-weighted annual collective dose based on more measurements than that ABE *et al.* had done in Osaka Pref.¹⁾ is important to obtain national collective dose in Japan. The Osaka Pref. mainly consists of granitic mountains on the east and the south sides and a wide alluvial plain in the center. The understanding of distributions of the dose rate due to geological conditions is fundamentally significant.

This paper describes the outdoor air absorbed dose rates measured at 40 sites by thermoluminescence dosimeters (TLDs), and relationships between the measured dose rates and the dose rate estimated by concentrations of U-series, Th-series and ⁴⁰K. Also measured were mineral compositions in the soil samples collected on the sites. The authors would suggest a simple method to estimate the dose rates based on the concentration of ⁴⁰K in soil samples.

II EXPERIMENT

1. Allotment of site

A total of 40 sites was selected to measure the absorbed dose rates in air in the Osaka Pref., as

shown in Fig. 1 and Table 1, in relation to geology, administrative boundary and population density. We allotted corners of playgrounds in 25 schools for the sites. The area of Osaka Pref. is 1866.86 km². Ryouke granitic and andesite mountains are located at the east and south sides of Osaka Pref. as shown in Fig. 2. The Osaka Pref. consists of Osaka city with a population of 2,000,000 and several satellite cities with a total population of several hundred thousands.

2. Absorbed dose rate in air

The TLD used in this study was a dosimeter containing two calcium sulfate chips.⁸⁾ Two sets of TLDs were put in a wooden box which were 1.2 m above the ground, and were kept at the sites for two months. The absorbed dose rates in air were obtained by averaging data of three cycles at same sites. Then, each absorbed dose rate in air is the mean over six months. The authors measured the absorbed dose rates in air at two sites for two years and noticed no seasonal effect. This may be caused by mild climate. In

Table 1 Sites where the absorbed dose rate in air and the concentration of radionuclides of soil were measured.

Site No.	Name of sites measured
1	Osaka Prefectural Misaki High School
2	Osaka Prefectural Sunagawa High School
3	Osaka Prefectural Sano High School
4	Osaka Prefectural Kishiwada High School
5	Osaka Prefectural Shinoda High School
6	Kaizuka City Sohara Primary School
7	Kaizuka City Third Junior High School
8	Kishiwada City Higashikatsuragi Primary School
9	Izumi City Makio Junior High School
10	Hagoromo Gakuen High School
11	Kawachi-Nagano City Takihata Primary School
12	Chihaya Akasaka Village Junior High School
13	Prefectural Tondabayashi Public Health Center
14	Prefectural Habikino Hospital
15	Minamikawachi County Taishi cho Taishi Junior High School
16	Pool Gakuin College
17	Osaka Prefectural Izumiootsu Public Health Center
18	Kinki Central Hospital
19	Osaka Prefectural Shuutokugakuin
20	Osaka Prefectural Yao Public Health Center
21	Osaka Prefectural Tennouji High School
22	Osaka Castle
23	Kawamata Water Disposal Plant
24	Osaka Prefectural Ichioka High School
25	Osaka Prefectural Moriguchi High School
26	Shijiyonawate City Shijiyonawate Minami Junior High School
27	Osaka Prefectural Kitano High School
28	Osaka Prefectural Kasugaoka High School
29	Neyagawa City Kunimatsu Primary School
30	Osaka Prefectural Murano Water Purification Plant
31	Osaka Prefectural Youth Recreation Center
32	Osaka Prefectural Takatsuki Minami High School
33	Osaka Prefectural Toyonaka Public Health Center
34	Osaka Prefectural Gardening High School
35	Minoo City Minoo Water Purification Plant
36	Health Center for Physically Handicapped Children
37	Osaka Prefectural Fukui High School
38	Osaka Prefectural Shiroyama High School
39	Osaka Prefectural Ranch
40	Osaka Prefectural Youth Field Exercise Center

order to eliminate unreliable sub-peak values and exclude fading in TLDs, the authors carried out preannealing of TLDs at 100°C for 4 min before reading the values as described in our paper.⁹⁾ The TLDs were calibrated by irradiating 87.3 μ Gy using ^{226}Ra and ^{137}Cs standard sources. We divided the whole of the Osaka Pref. into four sections and spent three years measuring the absorbed dose rates in air at all 40 sites.

3. Concentration of radionuclides and mineral composition

We collected soil samples of 5 cm thick core from surface of the ground to 10 cm or 30 cm depth by using an iron core sampler which is 9.5 cm in diameter and 30 cm in height. Each core sample of 5 cm thick was dried in air for several days and at 105°C in an oven for several hours and was sieved into two classes of particle sizes by using a standard sieve of 10 mesh. Soil particles smaller than 10 mesh were measured in that condition, but those larger than 10 mesh were crushed before their activities were measured. Each soil sample of 120 g was sealed into a polyvinyl chlorides case. Then each specimen was subjected to gamma-ray spectrometry with a planer type of Ge detector and a coaxial type of Ge(Li) detector. To reduce the background counting, the planer type of detector was surrounded with lead, brass and cadmium and the coaxial type of detector was placed in an iron shield with lead and polyvinyl chloride lining.¹⁰⁾ The activities of ^{238}U , ^{226}Ra , ^{228}Ra and ^{40}K were determined by the 63.3 keV gamma-rays of ^{234}Th , the 609 keV gamma-rays of ^{214}Bi , the 583 keV gamma-rays of ^{208}Tl and its 1.46 MeV gamma-rays, respectively. The authors used reference samples of NBL (the New Brunswick Laboratory of the Atomic Energy Commission) as a standard for the activity determination. In order to determine the mineral composition, each dried sample after desegregated in an agate mortar was subjected to X-ray diffraction analysis performed by using copper radiation and nickel filter.

4. Water content ratio

Water content ratio is a percentage value of net weight loss of a sample by drying divided by weight of fresh soil sample. Every fresh soil was kept sealed in a polyvinyl bag immediately after collecting and was weighed at first. The samples dried at 105°C as described above were weighed at second and, thus, the water content ratios were

Table 2 Dose rates in Osaka Pref.

Site number	Absorbed dose rate in air nGy/h	Exposure rate $\mu\text{R/h}$	Annual effective dose equivalent mSv/y
1	81.0 $\pm$ 0.3	9.28 $\pm$ 0.03	0.67 $\pm$ 0.01
2	94.4 $\pm$ 8.6	10.81 $\pm$ 0.98	0.76 $\pm$ 0.01
3	73.2 $\pm$ 1.7	8.38 $\pm$ 0.20	0.61 $\pm$ 0.00
4	81.0 $\pm$ 2.4	9.28 $\pm$ 0.27	0.67 $\pm$ 0.00
5	97.3 $\pm$ 1.2	11.14 $\pm$ 0.14	0.79 $\pm$ 0.00
6	85.8 $\pm$ 2.1	9.83 $\pm$ 0.24	0.70 $\pm$ 0.00
7	80.0 $\pm$ 1.8	9.16 $\pm$ 0.21	0.66 $\pm$ 0.00
8	104.1 $\pm$ 2.7	11.93 $\pm$ 0.31	0.83 $\pm$ 0.00
9	90.4 $\pm$ 2.1	10.35 $\pm$ 0.24	0.74 $\pm$ 0.00
10	85.5 $\pm$ 2.0	9.79 $\pm$ 0.23	0.70 $\pm$ 0.00
11	114.1 $\pm$ 1.0	13.07 $\pm$ 0.11	0.91 $\pm$ 0.00
12	67.0 $\pm$ 1.9	7.68 $\pm$ 0.22	0.57 $\pm$ 0.00
13	69.1 $\pm$ 1.0	7.92 $\pm$ 0.12	0.58 $\pm$ 0.00
14	75.9 $\pm$ 2.7	8.69 $\pm$ 0.31	0.63 $\pm$ 0.00
15	66.3 $\pm$ 2.3	7.60 $\pm$ 0.26	0.56 $\pm$ 0.00
16	80.6 $\pm$ 2.7	9.23 $\pm$ 0.31	0.67 $\pm$ 0.00
17	79.7 $\pm$ 2.4	9.13 $\pm$ 0.27	0.66 $\pm$ 0.00
18	74.0 $\pm$ 1.8	8.48 $\pm$ 0.21	0.62 $\pm$ 0.00
19	69.0 $\pm$ 2.7	7.90 $\pm$ 0.31	0.58 $\pm$ 0.00
20	71.5 $\pm$ 2.1	8.19 $\pm$ 0.24	0.60 $\pm$ 0.00
21	72.5 $\pm$ 1.6	8.30 $\pm$ 0.18	0.61 $\pm$ 0.00
22	79.8 $\pm$ 2.4	9.14 $\pm$ 0.27	0.66 $\pm$ 0.00
23	85.3 $\pm$ 0.3	9.77 $\pm$ 0.03	0.70 $\pm$ 0.01
24	84.6 $\pm$ 1.4	9.69 $\pm$ 0.16	0.69 $\pm$ 0.00
25	90.4 $\pm$ 1.7	10.36 $\pm$ 0.20	0.74 $\pm$ 0.00
26	89.5 $\pm$ 2.2	10.25 $\pm$ 0.25	0.73 $\pm$ 0.00
27	81.7 $\pm$ 1.2	9.36 $\pm$ 0.14	0.67 $\pm$ 0.00
28	82.5 $\pm$ 3.0	9.45 $\pm$ 0.34	0.68 $\pm$ 0.00
29	81.3 $\pm$ 1.8	9.31 $\pm$ 0.21	0.67 $\pm$ 0.00
30	88.3 $\pm$ 2.2	10.12 $\pm$ 0.25	0.72 $\pm$ 0.00
31	90.5 $\pm$ 1.7	10.37 $\pm$ 0.20	0.74 $\pm$ 0.00
32	108.3 $\pm$ 1.9	12.40 $\pm$ 0.22	0.86 $\pm$ 0.00
33	89.9 $\pm$ 1.5	10.30 $\pm$ 0.17	0.73 $\pm$ 0.00
34	75.6 $\pm$ 1.3	8.66 $\pm$ 0.15	0.63 $\pm$ 0.00
35	85.8 $\pm$ 1.3	9.83 $\pm$ 0.15	0.70 $\pm$ 0.00
36	80.3 $\pm$ 0.9	9.20 $\pm$ 0.11	0.66 $\pm$ 0.00
37	96.6 $\pm$ 1.2	11.07 $\pm$ 0.14	0.78 $\pm$ 0.00
38	80.9 $\pm$ 1.4	9.27 $\pm$ 0.16	0.67 $\pm$ 0.00
39	79.4 $\pm$ 1.7	9.10 $\pm$ 0.20	0.66 $\pm$ 0.00
40	101.5 $\pm$ 1.3	11.63 $\pm$ 0.15	0.82 $\pm$ 0.00

Table 3 Relationship between the main mineral compositions of the soils collected at 40 sites and the absorbed dose rates in air.

Group	Main mineral compositions	Site number	Absorbed dose rates in air nGy/h
Ⓐ	quartz, feldspar, mica, amphibole	2, 5, 6, 8, 9, 10, 11, 23, 24, 25, 26, 27, 29, 30, 31, 32, 33, 37, 40	81–115
Ⓑ	quartz, (feldspar)	1, 3, 4, 7, 18, 21, 22, 28, 35, 36, 39	72–86
Ⓒ	quartz, kaolinite, mica, amphibole, (feldspar, chlorite)	12, 13, 14, 15, 16, 17, 19, 20, 34, 38	66–81

Table 4 Dose rates in Osaka Pref.

The highest absorbed dose rate in air	114.1 ± 1.0 nGy/h
The lowest absorbed dose rate in air	66.3 ± 2.3 nGy/h
The central value of absorbed dose rate in air	81.0 ± 2.4 nGy/h
The mean value of absorbed dose rate in air	84.1 ± 0.4 nGy/h
Annual collective dose	5,198 man·Sv/y

determined.

III RESULTS AND DISCUSSION

Table 2 shows the air absorbed dose rates obtained by TLDs at all the 40 sites. **Figure 3** shows its frequency distribution. The soil particles smaller than 10 mesh collected at the 40 sites had mineral compositions as shown in **Table 3**. The soil samples were classified into groups of Ⓐ–Ⓒ as shown in **Table 3**. The group Ⓐ is mainly composed of soils originated from weathering of granite and the group Ⓑ consists of alluvial soil. The group Ⓒ is mainly composed of soils of Osaka group, andesite and basalts. Some samples of the Ⓑ and Ⓒ groups contain small amount of calcites. We obtained the highest value at the granite zone and the lowest value at the andesite zone as shown in **Table 4**. T. TSUJIMOTO *et al.*¹¹⁾ had determined air dose rates by an ionization chamber at the granitic zone as 12.0–14.2 $\mu\text{R/h}$ (105–124 nGy/h), at the Osaka group as 6.8–9.7 $\mu\text{R/h}$ (60–84 nGy/h) and at the alluvial zone as 8.5–10.7 $\mu\text{R/h}$ (74–94 nGy/h) in the Osaka Pref. The air absorbed dose rate obtained by our TLD measurements agree with the values of TSUJIMOTO *et al.* The absorbed dose rates in air at 1.2 m above the ground surface are given by the con-

Fig. 3 Frequency of the air absorbed dose rate measured by TLDs at 40 sites in Osaka Pref.

centrations of ^{238}U , ^{232}Th and ^{40}K in soil according to Eq. (1)¹²⁾ as described below; the calculations are based on the assumption that all decay products of ^{238}U and ^{232}Th are in radioactive equilibrium with their precursors. Open circles in **Fig. 4** show a relation between the air absorbed dose rates obtained by the TLD measurements (30 nGy/h is subtracted as cosmic radiation contribution) and the dose rates calculated by Eq. (1)¹²⁾ from the concentrations of ^{238}U , ^{238}Ra and ^{40}K in wet soil samples smaller than 10 mesh.

$$D = 0.044 \times K + 0.47 \times U + 0.63 \times Th \quad (1)$$

D : absorbed dose rate in air 1 m above the ground (nGy/h)

K : concentration of ^{40}K in wet soil (Bq/kg)

U : concentration of ^{238}U in wet soil (Bq/kg)

Fig. 4 The relation between the air absorbed dose rates obtained by subtracting cosmic radiation from the dose rates of TLDs measurements and those estimated by Eq. (1) from the concentrations of ^{238}U , ^{228}Ra and ^{40}K in samples smaller ($\circ$) and larger ($\bullet$) than 10 mesh.

Th: concentration of ^{232}Th in wet soil (Bq/kg)

We measured the concentrations of these three radionuclides in dried soil samples and then converted the concentrations into those per unit weight of wet soils by the use of the water content ratio of fresh soil. The dose rates estimated from soil activities are higher than those obtained by TLDs. Closed circles show the similar correlation in the case of soil particles larger than 10 mesh. The values of closed circles are slightly smaller than the values of open circles. This can be explained by the fact shown in Fig. 5. Figure 5 shows a correlation between the concentrations of ^{40}K and ^{228}Ra in soils smaller than 10 mesh and that in soils larger than 10 mesh. Concentrations of ^{40}K in soils larger than 10 mesh are higher than those in soils smaller than 10 mesh. On the contrary, the concentrations of ^{238}U , ^{226}Ra and ^{228}Ra in soils larger than 10 mesh are lower than those in soils smaller than 10 mesh. The reasons are as follows. As described in our previous paper,¹³⁾ for homologous soils resulting from progressive weathering, ^{238}U , ^{226}Ra and ^{228}Ra contents increase with an increase of surface area of soils as some of these nuclides are adsorbed on the surface area. However, ^{40}K contents decrease with an increase of surface area of soil as some of the ^{40}K is weathered away.

Figure 6 shows a relation between the air absorbed dose rates obtained by subtracting the cosmic radiation from the dose rate measured by TLDs and the estimated doses by the concentrations of all the soil particles irrespective of their

Fig. 5 The correlation between the concentration of ^{40}K and ^{228}Ra in samples smaller than 10 mesh (a) and that larger than 10 mesh (b).

Fig. 6 The relation between the air absorbed dose rates obtained by subtracting the cosmic radiation from the TLD measurements and those estimated by Eq. (1) from the concentrations of ^{238}U , ^{228}Ra and ^{40}K in all the samples irrespective of particle size.

size. As is shown, the rate, estimated from the whole samples are apparently close to the dose rates obtained by TLDs. However, some of the values estimated from the soils are higher than the dose rate obtained by TLDs. This can be explained as follows. The soils which had no stones larger than about 20 mm in diameter were collected as samples. While there were, in fact, some stones larger than about 20 mm in diameter in the ground, the dose estimated from the concentrations of the three nuclides in such stones

Fig. 7 A correlation between the air absorbed dose rates obtained by using TLDs and the concentration of ^{40}K in all of the samples irrespective of particle size.

were smaller than that of collected soil samples. In addition, there were some buildings in the vicinity. Furthermore, the surface soils at sites named A, B and C were carried from the granite zone when the school was built while the contents of ^{238}U , ^{226}Ra and ^{40}K in the soil deeper than 10 cm are lower than that of the surface soil.

A good correlation between the absorbed dose rates in air and the ^{40}K content in the soil was obtained as shown in Fig. 7. Then, we can roughly estimate the absorbed dose rate from the concentration of ^{40}K , that is easily obtainable, in the surface soil.

We estimated the population-weighted annual collective dose as $5,198 \text{ man}\cdot\text{Sv/y}$. This population-weighted annual collective dose is a little lower than $0.656 \times 10^6 \text{ man}\cdot\text{rad}$ ($5,380 \text{ man}\cdot\text{Sv}$) calculated by S. ABE *et al.*²⁾ This is because we calculated the value by using the absorbed dose rate in air at all the 40 sites containing andesite zone having a low population density, while S. ABE *et al.*¹⁾ calculated the value from the absorbed dose rates in air in the area having rather high population density in the Osaka Pref.

IV CONCLUSION

We obtained the central value of absorbed dose rates in air in the Osaka Pref. to be $81.0 \pm 2.4 \text{ nGy/h}$ by TLDs, and also obtained a fairly good correlation between the dose rates and concentrations of radionuclides in surface soils. It was found that the soil particles smaller than 10 mesh generally contribute more than that larger than 10 mesh to the dose rates, and it is anticipated that the dose rate can be estimated from concen-

trations of ^{40}K in surface soils.

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