ORIGINAL

Background Radioactivity of Soil in Wakasa Bay Area
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Soil samples of surface layer were collected in Wakasa Bay area which is composed of various geologic properties. Concentrations of background radionuclides in soil samples were determined by γ -ray spectrometry with a coaxial type and a planer type of Ge(Li) detectors. The spectra of soil samples obtained by X-ray diffraction analysis show the characteristic composition of minerals based on the geologic properties of their mother's rocks. The concentrations of ^{238}U , ^{226}Ra , ^{228}Ra and ^{40}K in soil roughly correspond to the geologic properties. The highest concentrations of these four nuclides are the soil collected from granitic zone and next comes the soil from paleozoic zone. For homologous soil from granite and paleozoic the ^{40}K contents decrease with increasing surface area, respectively. A good correlation between the ^{137}Cs contents and the unsupported ^{210}Pb contents was found in the soil samples of surface layer.

KEY WORDS: background radioactivity, soil, Wakasa Bay area, mineral composition, geologic properties, natural radioactivity

I INTRODUCTION

Knowledge of natural background radiation level is important not only in health physical research but in geoscientific research. It is well known that terrestrial radiation originates in crustal materials and natural environmental radiation varies with geology and geography.¹⁻³⁾ There have been many reports concerning distribution of natural radionuclides in rocks relating to their geologic properties¹⁻⁷⁾ and distribution of natural radionuclides in soil is usually assumed to be the same as that of their mother's rocks. A few investigations have been made concerning the distributions of natural radionuclides in relation to their

mineral compositions in soil over the ground.⁸⁻¹¹⁾ Detailed information about the distribution of natural radionuclides in soil is also necessary from a view point of an environmental radioactive monitoring.

Area of southern Fukui Prefecture which surrounds Wakasa Bay is composed of various geologic properties: granite, paleozoic, andesites, ultrabasic rock, yakuno intrusive rocks, alluvium. Radioactive fallout in this area, which is on the Japan Sea side, near the Asian continent, due to the old nuclear explosion tests deposited much compared with that on the side of the Pacific Ocean. The highest level of natural radiation in Japan was found in this area.¹⁾ A several nuclear power plants are constructed.

Soil samples were collected on fifty seven sites in this area and some investigations were made concerning the concentrations of uranium series nuclides (^{238}U , ^{226}Ra , ^{210}Pb), thorium series nuclides (^{228}Ra), ^{40}K and ^{137}Cs in soil in relation to their mineral compositions, their surface area and their ignition loss.

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II EXPERIMENT

The samples of soil were collected from the two layers of 0–5 cm and 5–10 cm on fifty seven sites in the zones of granite, paleozoic, andesite, ultrabasic rock, yakuno intrusive rock and alluvium classified in a geologic map as shown in Fig. 1. Soil samples were collected using an iron sampler 8 cm in a diameter and is dried in an oven at 110°C for 24 hr. The part of the sample of larger than 10 mesh is excluded by sieving and 120 g of dried sample are packed in a disk-shaped plastic case 9 cm in diameter and 2 cm in height. Each sample is subjected to γ -ray spectrometer with a planer type Ge(Li) detector (16 mm in diameter and 5.13 mm in thickness) and a coaxial type of Ge(Li) detector (effective volume 50 ml). In order to reduce a background counting rate the Ge(Li) detectors are surrounded with shielding materials.⁹⁾ The activities of ^{238}U and ^{210}Pb are determined by using the planer type Ge(Li) detector from photopeaks due to 63.1 keV of ^{234}Th (daughter nuclide of ^{238}U) and its 46.5 keV, respectively. (Even if uranium is chemically separated, 10 months is long enough to recover 99.9% of the equilibrium between ^{238}U and ^{234}Th (half-life 24.1 days)). The activity of ^{226}Ra , ^{228}Ra (^{228}Ac) and ^{40}K are determined by using the coaxial Ge(Li) detector from the photo-

peaks due to 609 keV of ^{214}Bi , 911 keV and 1.46 MeV, respectively. The ^{137}Cs is determined from the photopeak due to 662 keV of $^{137\text{m}}\text{Ba}$. A reference sample by NBL (the New Brunswick Laboratory of the Atomic Energy Commission) is used as the standard for the activity determination.

X-ray spectra are measured for each dried sample after disaggregating in an agate mortar. Powder X-ray diffraction analysis is performed using copper radiation and nickel filter to determine mineral compositions.^{12,13)}

The surface area of soil particles is determined by rapid surface area analysis using low-temperature nitrogen adsorption.¹⁴⁾

Each sample of ignition loss is determined from the percentage of loss in weight after that the dried sample of 1 g is kept at 1,000°C for 1 hr¹⁵⁾ and an ignition loss is considered as a content of organic matter.

III RESULT AND DISCUSSION

1. Mineral composition

Figure 2 shows a typical X-ray diffraction spectrum of soil sample collected from zones of granite, paleozoic, yakuno intrusive rocks, ultrabasic rocks and andesine classified on a geologic map, respectively. Peaks due to quartz, feldspar, illite, kaolinite and chlorite are seen in a spectrum

Fig. 1 Geologic map of the Wakasa Bay area showing the locations of sampling site.

Fig. 2 Typical X-ray diffraction spectra of the soil samples collected from the zones of granite, paleozoic, yakuno intrusive rocks, ultrabasic rocks and andesine classified on a geologic map.

of soil collected from granite zone on the map. As the samples of soil from paleozoic zone were well weathered, clear peak due to quartz which is resistance to weathering and peaks due to illite, chlorite and kaolinite are shown in the spectrum of soil described above. Peaks due to pyroxene, chlorite, termolite, quartz are seen in a spectrum of soil collected from a zone of intrusive rocks. Ultrabasic rock is affected by desilication and peaks due to kaolinite and quartz are shown in its spectrum of soil. The peaks due to crystobalite and maghemite are also seen in that of soil collected from zone of andesites.

2. Natural radioactivity

Relationship between the concentrations of ^{238}U and ^{226}Ra , ^{226}Ra and ^{228}Ra are shown in Fig. 3. An arrangement in a descend order of the concentrations of these nuclides roughly are that of soil from granite, paleozoic, andesites, yakuno intrusive rocks and ultrabasic rocks. The ^{226}Ra activity is approximately equal to the ^{238}U activity in the soil samples except a few soil samples from granite. A soil sample containing 5.1 pCi/g of ^{238}U contains highly mica in which uranium is well known to be maldistributed.¹³⁾ A few soil samples con-

taining 3–4 pCi/g of ^{238}U were collected from vegetable field and paddy field and these rather high contents of uranium are due to chemical fertilizer.¹⁵⁾

The activity ratio of ^{228}Ra to ^{226}Ra is nearly unity in the soil samples except that from granite. These ratios are smaller than unity in two soil samples for the high uranium contents as described above. The activity ratios of ^{228}Ra to ^{226}Ra in the soil samples from granite are larger than unity and this reason seems that uranium which is a precursor of ^{226}Ra moves relative to thorium which is a precursor of ^{228}Ra .

The radioactive relationships between ^{40}K and ^{238}U or ^{228}Ra are shown in Fig. 4. The samples of two which show the highest values in the concentration of ^{40}K content clearly orthoclase. The samples from granite show nearly the same value as the samples from the others in the activity ratios of ^{238}U ($\cong$ ^{226}Ra as shown in Fig. 3(a)) to ^{40}K and, however, the activity ratio of ^{228}Ra to ^{40}K in the sample from granite is larger than those of samples from others. It is considered that ^{228}Ra contents in soil samples from granite are high compared with the ^{238}U , ^{226}Ra and ^{40}K contents.

(a)

(b)

Fig. 3 Relationship between the concentrations of ^{238}U and ^{226}Ra , ^{226}Ra and ^{228}Ra in the soil samples collected from the two layers of 0–5 and 5–10 cm.

This fact may also be based on ^{232}Th which is a precursor of ^{228}Ra .

We had reported that for an homologous soil, resulting from progressive weathering, which is free from the clay consisting of three-layer units, the ^{40}K contents decrease with increasing their surface area.¹⁰⁾ As shown in Fig. 5, for the soil samples which hardly content the clay of montmorillonids and illites, the ^{40}K contents in samples collected from the zones of granite and paleozoic decrease with increasing their surface area, respectively. The fact described above corresponds to the fact that the alkali metal ions and those of the alkaline earths are for the most part carried away in solution by erosion.¹³⁾

(a)

(b)

Fig. 4 The contents of ^{238}U and ^{228}Ra as a function of the ^{40}K contents in the soil samples collected from the two layers of 0–5 and 5–10 cm.

3. Relation between the ^{137}Cs content and ^{210}Pb content

The ^{137}Cs and ^{210}Pb contents in the samples of two soil layers in the depth of 0–5 cm and 5–10 cm are nearly equivalence for soil samples collected in vegetable and rice fields and sandy beaches, because of mixing of the two layers. For the samples collected in lots grown with bamboo, grass or turf, these contents in the samples of down layer are

Fig. 5 Concentrations of ^{40}K as a function of the surface area of soil collected from the zones of granite and paleozoic classified on a geologic map.

rather high because of that solution containing these nuclides flow down-wards with their roots. These concentrations in the two soil layers of samples except the described above are different from each other.

In Fig. 6 is shown a relationship between the ^{137}Cs contents and the contents of unsupported ^{210}Pb , i.e., ^{226}Ra supplied ^{210}Pb has been subtracted from the total ^{210}Pb . Some part of gaseous ^{222}Rn produced from ^{226}Ra in soil escapes to the air and the content of ^{210}Pb which is a decay product of ^{222}Rn may be a little bit less than the ^{226}Ra content. However, the contents of the total ^{210}Pb are mostly high compared with the difference between the ^{226}Ra contents and the contents of its supplied ^{210}Pb . Then radioactivity equilibrium between the

contents of ^{226}Ra and ^{210}Pb in soil is assumed to have been established and the content of unsupported ^{210}Pb is determined by the subtraction of the ^{226}Ra content from the total ^{210}Pb . It is considered that ^{222}Rn exhaled from the ground and its decay products stay in the atmosphere and deposit on the ground with a rainout and a dry fallout. ^{137}Cs which is produced by the nuclear explosion tests also deposits on the ground with a rainout and a dryout. In this figure is also shown the values of the contents in the samples collected from the place on which the fallout depositions are easily concentrated. It is observed that for the soil samples collected in 1881–1983 from the two layers in the depth of 0–5 and 5–10 cm, a relation in activities of the ^{137}Cs to the unsupported ^{210}Pb may be expressed by the following formula;

$$C = 0.29B - 0.24$$

C : the ^{137}Cs content (pCi/g)

B : the unsupported ^{210}Pb content (pCi/g)

This correlation may be inapplicable for the soil samples collected after several years, because of that unsupported ^{210}Pb always deposits and, however, ^{137}Cs almost deposited until 1972 and migrates downwards in the ground, and the ^{137}Cs content in soil decreases with year according to its half life also. This relation in activities of the two nuclides is mainly based on the datum about soil enriched in organic matter and then, these two nuclides are mainly considered to associate with organic matter.^{16,17)} For estimating influence of an accident of nuclear energy plants, the levels of ^{137}Cs in soil are generally monitored. The contents of ^{137}Cs in the soil of 0–10 cm layer due to nuclear explosion tests can be estimated by this formula

Fig. 6 A relationship between the ^{137}Cs contents and the contents of unsupported ^{210}Pb in the soil samples collected from the two layers of 0–5 and 5–10 cm.

with the unsupported ^{210}Pb . The levels of ^{137}Cs due to an accident of nuclear energy plant may be estimated.

4. Ignition loss and the ^{137}Cs contents and the ^{210}Pb contents

In Fig. 7 is shown a relationship between ignition loss and the ^{137}Cs contents and the ^{210}Pb contents, respectively. These contents have a tendency to increase with increasing of the ignition loss of sample for an each case of places having a similar value in the total depositions.^{16,17} The following three places; the place of under trees is covered with dead leaves collected the fallout radioactivity,

Fig. 7 The ^{137}Cs contents and the ^{210}Pb contents as a function of ignition loss.

Fig. 8 A distribution of the concentrations of ^{40}K , ^{238}U , ^{226}Ra , ^{228}Ra , ^{210}Pb and ^{137}Cs in the soil samples collected from the two layers.

Fig. 9 The frequency of the concentrations of ^{238}U , ^{226}Ra , ^{210}Pb , ^{228}Ra and ^{137}Cs in the soil samples of 0-5 cm layer.

as rainout and dryout are adsorbed very much on surface of leaves; as rain water also carry fallout radioactivity, on place which rain water is easily gathered; the last place is bare land.

5. Radioactivity of soil

As shown in Fig. 8, a distribution of the ^{40}K content is the widest in the contents of background radioactive nuclides. It is seen in Fig. 9 that the radioactive concentration of ^{210}Pb , which is a decay product of radon, is the highest in the background radioactivities of soil except that of ^{40}K .

IV REMARKS

It was found that mineral compositions of soil are based on the geologic properties of their mother's rocks and the contents of ^{238}U , ^{226}Ra , ^{228}Ra and ^{40}K in soil are characterized in geologic properties. For homologous soil from granite and paleozoic the ^{40}K contents decrease with increasing surface area, respectively. It is observed that the radioactive concentration of ^{210}Pb is the highest of background radioactivity in soil except that of ^{40}K . A good correlation between the ^{137}Cs and the unsupported ^{210}Pb contents was found in soil samples in the depth of 0–10 cm and then, the activity of ^{137}Cs due to an accident of nuclear energy plants may be easily distinguish from that due to nuclear explosion tests using this correlation.

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