

VERTICAL DISTRIBUTION OF NATURAL RADIOACTIVE NUCLIDES IN SOIL  
(DEPTH OF 0 TO 30 CM)

Kazuko MEGUI\*, Shojiro KIMURA\*\*, Tadashi TSUJIMOTO\*\*\* and Kousuke KATSURAYAMA\*\*\*

\*:Rad.Ctr.Osaka Pref., \*\*:Osaka Univ. Pharm.Sci., \*\*\*:Res.Reac. Inst. Kyoto Univ., \*\*\*\*:Kinki Univ.

INTRODUCTION

Knowledge of vertical distribution of natural radioactive nuclides in soil is very important in health physics. Works concerning the vertical distribution of the artificial nuclides in soil have been widely studied. Few studies were reported on the vertical distribution of a several natural radionuclides in soil and, however, these distribution are considered to be important in investigation of their mobility in soil. The study reported in this paper presents data on the distribution of U-238, Ra-226, Pb-210, Ra-228 and K-40 in soil core samples.

SAMPLING AND MEASUREMENT

Soil core samples of 30 cm depth in length were taken on the ground in Fukui Pref. and Osaka Pref., which is located in the central part of Japan, and the former is on the Japan Sea side and the latter is on the side of the Pacific Ocean. The area collected the samples is composed of various geologic properties. Core samples of soil were collected by using an iron sampler. Each core sample of 30 cm long was sliced into 5 cm section and dried. The part of the soil sample of larger than 10 mesh is excluded by sieving and 120 g of dried sample are packed in a disk-shaped plastic case.

The soil sample are examined in the concentration of U-238, Ra-226, Pb-210, Ra-228 and K-40 by gamma-ray spectrometry with a coaxial and a planer type of Ge detectors. To reduce the background counting rate, the detectors were surrounded with the shielding materials<sup>(1)</sup>. The activities of U-238 and Pb-210 are determined by using the planer type Ge detector from photopeaks due to 63.3 keV of Th-234 (daughter nuclide of U-238) and its 46.5 keV, respectively. The activities of Ra-226, Ra-228 and K-40 are determined by using the coaxial type Ge detector from the photopeaks due to 609 keV of Bi-214, 911 of Ac-228 and 1.46 MeV, respectively. A reference sample by NBL (the New Brunswick Laboratory of the Atomic Energy Commission) is used as the standard for the activity determination.

The Fe, Mn and Ca contents in samples were determined by atomic absorption spectrometry according to Bruland et al.<sup>(2)</sup> and Terashima<sup>(3)</sup>. The soil sample of 1.0 g was decomposed with a mixture of HF and HCl<sub>4</sub>. After having been converted to the chlorides by HCl, the molecular absorption interferences were suppressed by addition of strontium.

RESULT AND DISCUSSION

As shown in Fig. 1, no distinct vertical difference was found in the concentration of U-238, Ra-226 and Ra-228 in the soil samples except a single case described afterward. The concentrations of K-40 in the soil samples were found to decrease with increase in ignition loss of soil samples, as shown in Figs. 1 and 2. The slightly low concentrations of K-40 in the top layer were usually found compared with those in the deeper layers of soil. This may be caused by the fact that the alkali metal and those of the alkaline earths in the soil sample of the top layer easily carried away in solution by erosion.

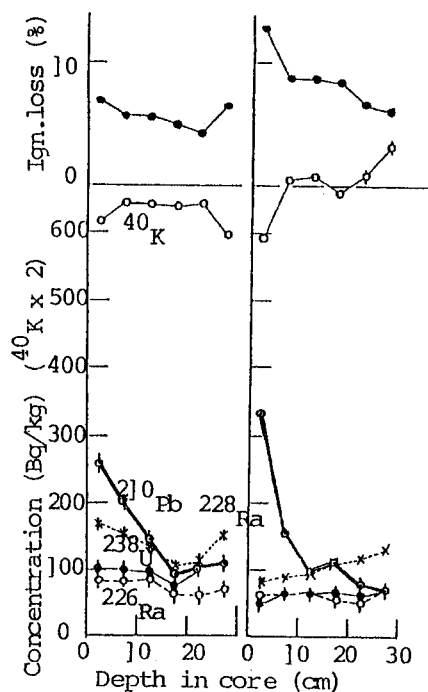


Fig.1 Concentrations of U-238, Ra-226, Pb-210, Ra-228 and K-40 and ignition loss as a function of depth in core for the first case.

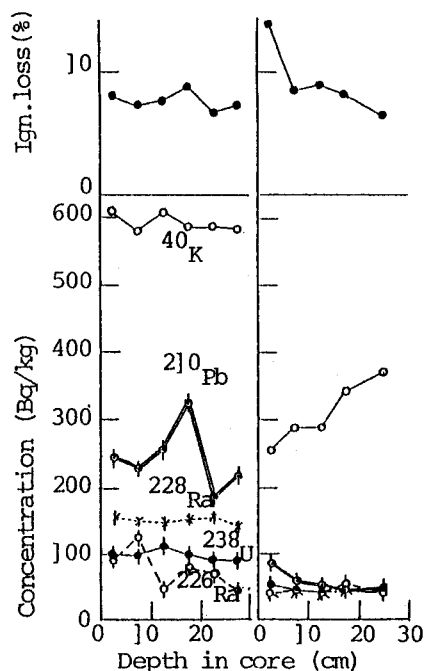


Fig.2 Concentrations of U-238, Ra-226, Pb-210, Ra-228 and K-40 and ignition loss as a function of depth in core for the third case.

The vertical variations of the Pb-210 contents in soil might be classified into four cases. For the first and the second cases of vertical distributions of Pb-210, the Pb-210 contents were the highest at the top layers (0-5 cm) and decrease with the depth. As shown in Fig. 1 for the first case, the concentration of Pb-210 in the 25-30 cm layers was established radioactive equilibrium with the concentrations of U-238 and Ra-226. For the second case, the Pb-210 content in the 25-30 cm layer was higher than the contents of U-238 and Ra-226 in the layers. The liquid phase contained Pb-210 permeates into the deeper part of 30 cm depth. For the third case, the Pb-210 contents were almost uniformly, as shown in Fig. 2, because of artificial mixing of soil layers. For these three cases described above a rather good relationship was found between the Pb-210 contents and the organic contents.

Fig.3 shows the vertical distribution of U-238, Ra-226, Pb-210, Ra-228 and K-40 on the the last case which is singular. The concentrations of U-238, Pb-210 and Ra-228 in the soil layer show the highest values in the 10-15 cm layer and the Mn and Fe contents are also high in the same layer. Fig. 4 shows the

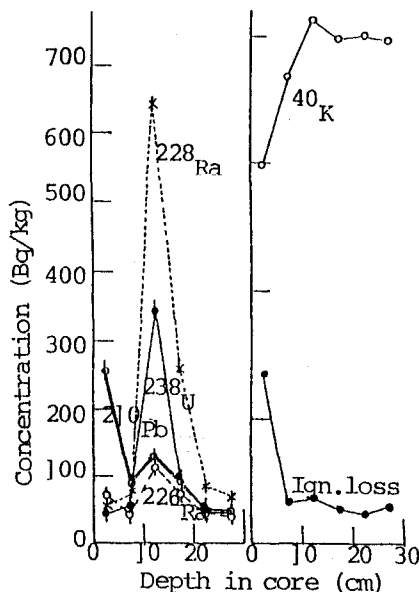


Fig. 3 Concentration of U-238, Ra-226, Pb-210, Ra-228 and K-40 and ignition loss as a function of depth in core for the last case.

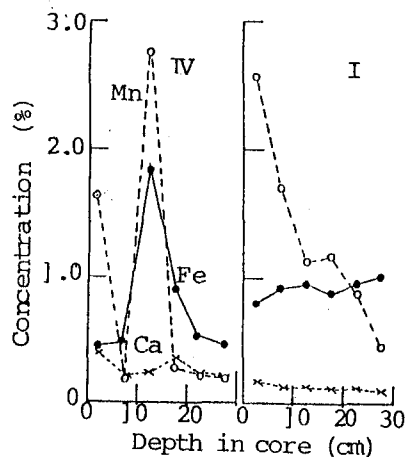


Fig. 4 Concentration of Fe, Mn and Ca as a function of depth in core.

the variations of these contents on the last case and the first case which is the most general case. In the general case the Mn contents are the highest in the 0-5 cm layer and gradually decrease with the depth, and the Fe and Ca profiles in both cores show relative uniformity with depth. This is due to the fact that manganese dissolve in reduced layer, then slowly migrate and accumulates in the oxidized top layer. Iron forms insoluble compounds in the oxidized and the reduced layers. No appreciable change was found in the K contents. A possible explanation of relatively high concentration of U-238 and Ra-228 is as follows. The organic compounds in the top layer decompose and form humic and fulvic acids in the oxidized conditions. Metal elements are complexed with fluvic acid and transported downward in solution. And then, the metal complexes accumulate when the ratio of metal and fulvic acid attain to a constant value. Thus, uranium and thorium show relatively large accumulations in 10-15 cm deep.

The migration rate of Pb could be determined by evaluating vertical decrement with depth in the Pb-210 contents and its half-life for the first case described above, as shown in Fig. 5. The migration rate of Pb-210 were lead to the values of 0.2 in an usual area and 0.36 mm/yr in a moisture area.

Table 1 is shown the total accumulation of Pb-210 on the ground and yearly rain fall at Wakasa and Sakai,

which the former is on the Japan Sea side, near the Asian Continent and the latter is on the side of the Pacific Ocean. Then, the total accumulation at Japan Sea side is several times higher than that at the side of the Pacific Ocean due to influence of the Asian Continent.

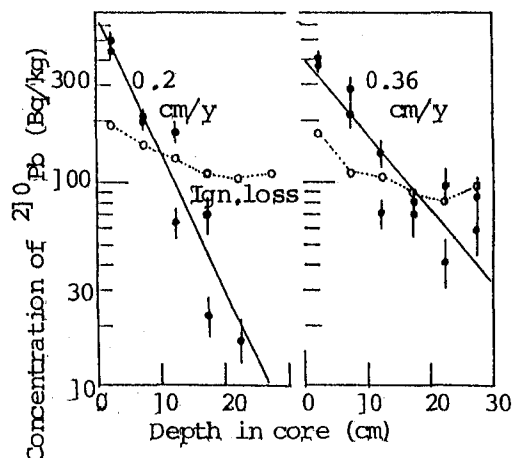


Fig.5 Migration rate of the surface soil layer collected in Wakasa.

Table 1 Total accumulation of Pb-210 and yearly rainfall in Wakasa and Sakai

Location	Total accumulation of Pb-210 <sub>2</sub> (kBq/km <sup>2</sup> )	Yearly rainfall (mm/y)
Wakasa	20.2	2560
	21.3	
	29.1	
	21.4	
Sakai	2.6	1390
	2.3	

#### CONCLUSION

The vertical distribution of the Pb-210 contents in the surface soil layer might be classified into four cases. The migration rate of Pb-210 could be determined by evaluating vertical decrement with depth in the Pb-210 contents and were lead to values of 0.2 in an usual area and 0.36 mm/yr in a moisture area. The concentration of U-238, Pb-210 and Ra-228 in the soil layers showed the highest values in the 10-15 cm layer and the Mn and Fe contents were also high in the same layer and, however, this is singular.

**ACKNOWLEDGMENT** The authors wish to express their gratitude to Dr. S. Abe of National Institute of Radiological Sciences, Dr. Y. Nakashima of Nagoya University and T. Ishiyama of Radiation Center of Osaka Pref. for their useful advice. The authors are also indebted to Dr. K. Yamazaki, I. Urabe, K. Okamoto and T. Yoshimoto for useful discussions and collecting samples.

#### REFERENCES

- 1) K.Megumi and T.Mamuro, J. Geophys. Res., 82,353(1977)
- 2) K.W.Bruland, K.Bertin, M.Koide and E.D.Goldberg, Environ. Sci. Technol., 8, 425(1974)
- 3) S.Terashima, Jap. Anal. 19, 1197(1970)