

NATURAL AND ARTIFICIAL RADIONUCLIDES IN  
ENVIRONMENTAL SAMPLES COLLECTED FROM  
THE MCMURDO SOUND REGION IN SOUTH  
VICTORIA LAND, ANTARCTICA

Kazuhisa KOMURA\*, Tetsuya TORII\*\*, Masayoshi YAMAMOTO\*\*\*  
and Masanobu SAKANOUÉ\*

\*Low Level Radioactivity Laboratory, Kanazawa University,  
Wake, Tatsunokuchi-machi, Nomi-gun, Ishikawa 923-12

\*\*Chiba Institute of Technology, Tsudanuma, Narashino 275

\*\*\*Faculty of Pharmacy, Hokuriku University, Kanagawa-machi, Kanazawa 920-11

**Abstract:** Natural and artificial radionuclides in the soils collected from Ross Island and the Dry Valleys area and water filters used at the Scott Base have been studied by non-destructive  $\gamma$ - and  $\alpha$ -spectrometry. The U, Th and K contents in the soils from Cape Royds were found to be several times higher than those from the Dry Valleys area, while the K/Th and U/Th ratios were nearly the same independent of the sampling location. Accumulation of  $^{137}\text{Cs}$  in surface soils was estimated to be 1-5 mCi/km<sup>2</sup>, which is one to two orders of magnitude lower than those measured for the surface soils in Japan. In spite of the great difference of the fallout accumulation in surface soil, the  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio (0.016-0.022) was almost the same as the global fallout value. On the other hand, various artificial radionuclides were detected in the water filter samples. Among them, existence of  $^{207}\text{Bi}$  is remarkable in the Antarctic samples. No appreciable difference from the global fallout value was found for the  $^{239,240}\text{Pu}/^{137}\text{Cs}$  and  $^{241}\text{Am}/^{239,240}\text{Pu}$  activity ratios. However, the  $^{235}\text{Pu}/^{239,240}\text{Pu}$  ratio was found to be 3 to 5 times higher than those measured in the northern hemisphere, which can be attributed to the accidental release of  $^{235}\text{Pu}$  by the burn-up of SNAP-9A satellite in 1964.

## 1. Introduction

Antarctica is believed to be one of the most clean areas on the earth. However, contamination by various kinds of pollutants including artificial radionuclides has been gradually increasing due to the human activities. Recent increase of atomic power plants has enforced us to measure environmental radioactivities. Measurement of radioactivity in environmental samples is of interest from the viewpoint not only of environmental pollution, but also of meteorological and geochemical aspects.

Natural and artificial fallout nuclides in the snow and the ice sheet and also in the atmosphere of the polar region have been measured for the estimation of the rate of snow accumulation (PICCIOTTO and WILGAIN, 1963; PICCIOTTO *et al.*, 1964; CROZAZ *et al.*, 1964; LANGWAY *et al.*, 1965; OESCHGER *et al.*, 1966; CLAUSEN, 1973) and for the environmental assessment (MARTELL, 1959; LOCKHART *et al.*, 1965; FEELY *et al.*,

1980). However, data on the environmental radioactivities in Antarctica are rather few as compared with the abundant data for other areas, and this is primarily due to the limitation of available samples.

In the present work, we report the measurements of natural radionuclides belonging to U and Th series and  $^{40}\text{K}$ , and artificial radionuclides including  $^{137}\text{Cs}$ , plutonium isotopes,  $^{241}\text{Am}$  in the soils from the McMurdo Sound region of Southern Victoria Land and the water filters used at the Scott Base in Antarctica.

## 2. Samples

In order to measure the level of natural radioactivities and to estimate the contamination of Antarctica by artificial radionuclides, ground surface soils were collected in the 1979/80 summer season from various locations in the McMurdo Sound region. Strictly speaking, there may be no 'soil' in Antarctica, but in this paper, we use the term 'soil' for unconsolidated ground surface materials.

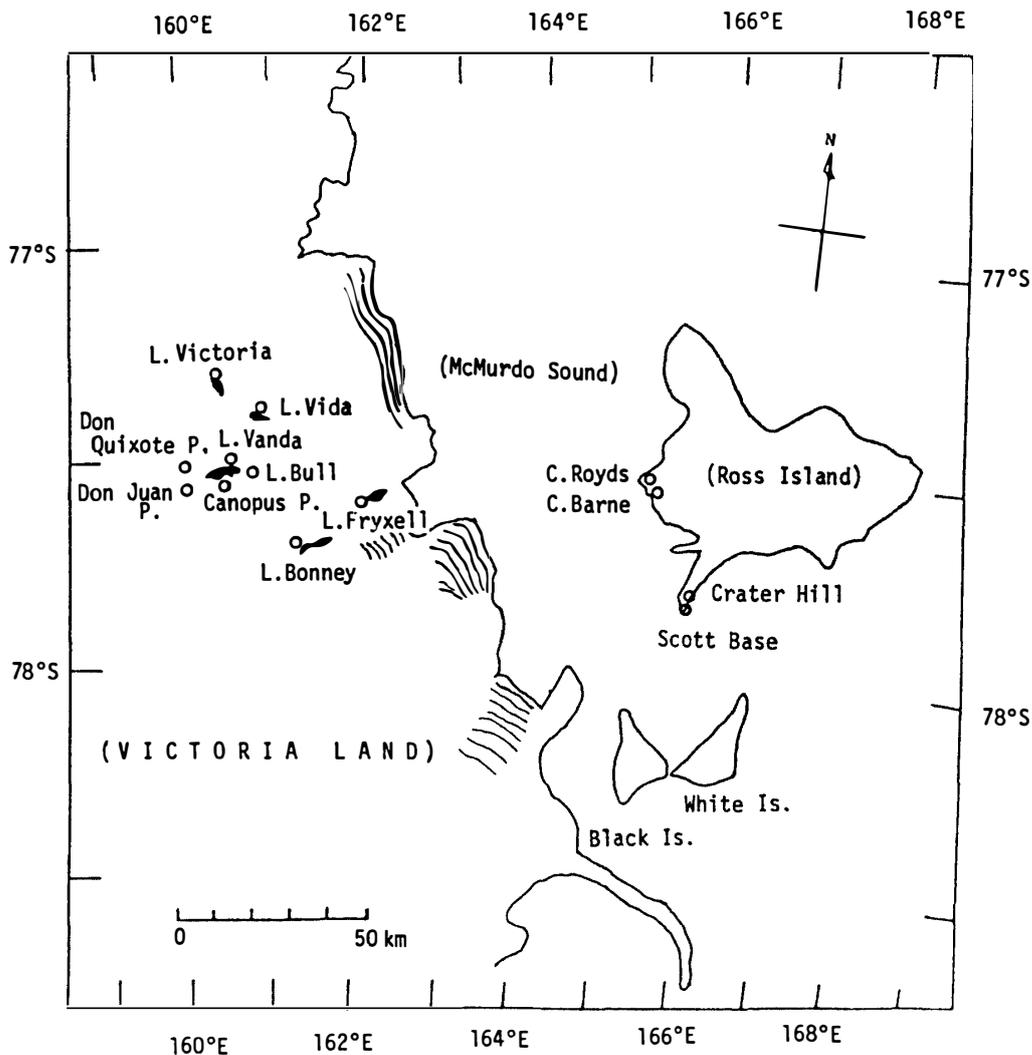


Fig. 1. Sampling locations of surface soils in the McMurdo Sound region, Antarctica.

The sampling sites of surface soils are shown in Fig. 1. In order to know the depth profile of radionuclide and to estimate the fallout accumulation, a flat area was chosen for the sampling site, and the surface soil down to 10–20 cm in depth was collected at intervals of several centimeters using a small scoop. Area of sampling was usually  $10 \times 10$  cm. Where the soil is sandy and homogeneous, a surface layer within 5 cm in depth was collected. The soil sample at Lake Fryxell was collected from the lichen-grown area near the terminus of the Canada Glacier, since the concentration of fallout nuclides in lichen tissue is known to be higher than that of bare soil (HOLM and PERSSON, 1976) and suitable to measure the fallout nuclides in low contamination area such as Antarctica. Since a fair amount of organic matter was contained in the 0–1 and 1–2 cm portions, it was decomposed by ashing in the electric furnace at  $450^\circ\text{C}$  before preparing the counting source.

Each soil was air-dried at room temperature and about 50 to 150 g of sample was taken for the non-destructive  $\gamma$ -spectrometry. The counting source was prepared simply by packing the sample in a plastic case of 60 mm in diameter or by compressing it into a 50 mm disc using a hydraulic press.

In order to study the fallout radionuclides in the ice sheet, water filters used at the Scott Base for the preparation of clean water were available through the courtesy of the Scott Base of New Zealand Antarctic Research Programme. Since particulate matter included in tons of meltwater can easily be sampled by the filtration, the used water filter is considered to be one of the most suitable materials to measure the artificial radionuclide in Antarctica. According to the scientists of the Scott Base, the snow and/or ice used for the preparation of water was taken from the area called 'Cliff' near the Scott Base, and each filter unit, which has the dimensions of  $7 \text{ cm}\phi \times 25 \text{ cm}$  and is made of a vegetable fiber string tightly coiled around the core post of 3 cm in diameter, was used to purify about 2–3 tons of meltwater.

In order to measure the radionuclides effectively, each filter was ashed at  $450^\circ\text{C}$  in an electric furnace. By ashing, the sample weight was reduced to only 2 to 10 g depending on the amount of particulate matter. The counting source of 35 or 50 mm in diameter was prepared by compressing the ash by means of a hydraulic press.

### 3. Measurement

The counting sources thus prepared were measured non-destructively by using two types of low background germanium detector. The first one is an ordinary Ge(Li) detector with 1.8 keV of energy resolution (FWHM) and 16% efficiency relative to  $7.6 \text{ cm}\phi \times 7.6 \text{ cm}$  NaI (TI) detector at 1.33 MeV, and the other is a Ge-LEPS (low energy photon spectrometer) with  $3.2 \text{ cm}\phi \times 1 \text{ cm}$  of Ge crystal and 0.5 keV of energy resolution at 59 keV. Each counting source was set on the end-window of the detector and the  $\gamma$ -ray spectrum was measured for one to several days.

After the non-destructive  $\gamma$ -spectrometry, most of the water filters and some soils were further subjected to the  $\alpha$ -spectrometry to measure artificial Pu isotopes and  $^{241}\text{Am}$  and natural U isotopes. The Th isotopes were also measured for some water filters. These elements were extracted from the sample by treating with mixed acid of  $\text{HNO}_3$  and  $\text{HCl}$  and also with  $\text{HF}$ . Known amount of  $^{232}\text{U}$  equilibrated with its

daughter  $^{228}\text{Th}$ ,  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  were initially spiked to the sample solution as the yield tracers of these elements during the chemical procedures. The Th, U, Pu and Am isotopes in the sample were pre-concentrated by co-precipitation with  $\text{Fe}(\text{OH})_3$  and then separated and purified systematically by the ion exchange method using anion and cation exchange resin columns. Finally, thin and uniform counting sources were prepared by electrodeposition onto stainless steel discs of 25 mm in diameter. The  $\alpha$ -ray spectrum of each counting source was measured for several days by using a surface-barrier type Si(Au) detector with 450 mm<sup>2</sup> of active area. Measured nuclides are listed in Table 1 together with their decay properties and possible origin.

Table 1. Measured radionuclides and their origin.

Nuclide	Half-life	Measured radiation	Origin and producing reaction
[Natural radionuclides]			
$^{40}\text{K}$	$1.28 \times 10^9$ y	1461 keV $\gamma$	Natural potassium
$^{210}\text{Pb}$	22.26 y	46 keV $\gamma$	Fallout, daughter of airborne $^{222}\text{Rn}$ uranium series nuclide
$^{230}\text{Th}$	$7.52 \times 10^4$ y	4.68 MeV $\alpha$	Airborne dust, uranium series nuclide
$^{232}\text{Th}$	$1.39 \times 10^{10}$ y	4.01 MeV $\alpha$	Airborne dust, parent of thorium series
$^{234}\text{U}$	$2.44 \times 10^5$ y	4.78 MeV $\alpha$	Airborne dust, uranium series nuclide
$^{238}\text{U}$	$4.46 \times 10^9$ y	4.20 MeV $\alpha$	Airborne dust, parent of uranium series
$^7\text{Be}$	53.4 d	478 keV $\gamma$	Induced by cosmic-ray reactions on air component
[Artificial radionuclides]			
$^{54}\text{Mn}$	312 d	835 keV $\gamma$	Neutron induced reaction on bomb material
$^{60}\text{Co}$	5.27 y	1332 keV $\gamma$	Neutron induced reaction on bomb material
$^{125}\text{Sb}$	2.77 y	428 keV $\gamma$	Fission product
$^{134}\text{Cs}$	2.06 y	570 keV $\gamma$	Fission product
$^{137}\text{Cs}$	30.0 y	662 keV $\gamma$	Fission product
$^{144}\text{Ce}$	284 d	133 keV $\gamma$	Fission product
$^{155}\text{Eu}$	4.96 y	105 keV $\gamma$	Fission product
$^{207}\text{Bi}$	38 y	1064 keV $\gamma$	Neutron induced reaction on bomb material
$^{235}\text{Pu}$	87 y	5.50 MeV $\alpha$	Neutron induced reaction on $^{239}\text{Pu}$
$^{239}\text{Pu}+$	24082 y	5.15 MeV $\alpha$	Raw material of nuclear bomb, most of
$^{240}\text{Pu}$	6538 y	5.16 MeV $\alpha$	$^{240}\text{Pu}$ is produced by neutron capture
$^{241}\text{Am}$	433 y	5.49 MeV $\alpha$	Daughter of $^{241}\text{Pu}$ produced by multiple neutron capture reaction on $^{239}\text{Pu}$

## 4. Result and Discussion

### 4.1. Surface soils

Radionuclides in more than 40 soil samples collected from various locations in the McMurdo Sound region have been measured by non-destructive  $\gamma$ -spectrometry. Depth profiles of U, Th, K and fallout nuclides were studied for the soil samples from Cape Royds and Crater Hill in Ross Island, Don Juan Pond, Don Quixote Pond and Canopus Pond in the Wright Valley and Lake Fryxell in the Taylor Valley.

Example of  $\gamma$ -ray spectrum is shown in Fig. 2 for the 0–2 cm layer of the soil at Cape Royds. As shown in Fig. 2, major  $\gamma$ -ray peaks are attributed to natural radio-

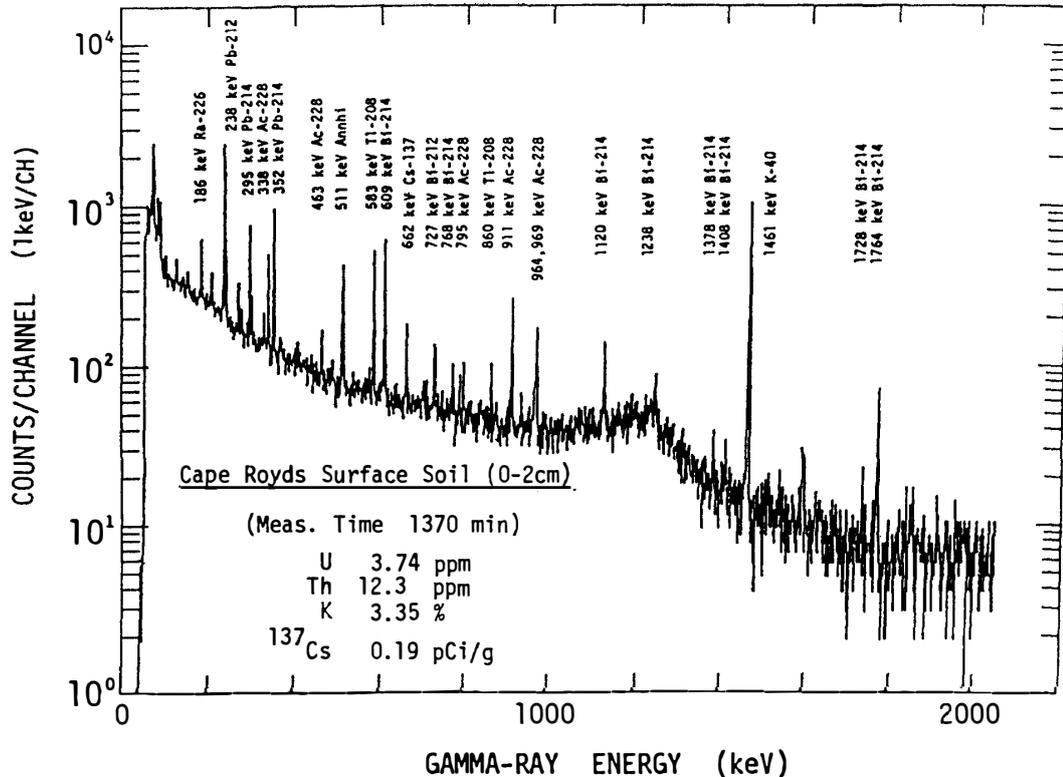


Fig. 2. Ge(Li) spectrum of surface soil collected from Cape Royds in Ross Island, Antarctica.

nuclides belonging to U and Th series and  $^{40}\text{K}$ , and the contribution of artificial  $^{137}\text{Cs}$  is rather small as compared with that of natural radionuclide. Except  $^{137}\text{Cs}$ , no artificial radionuclide was detected in the Ge(Li) spectrum. Measurement of plutonium isotopes was performed only for the soils containing detectable amount of  $^{137}\text{Cs}$ .

Results of measurements are summarized in Tables 2 and 3. Errors cited in the tables consist only of the  $1\sigma$  error of the counting statistics and no other source of error is accounted. The U and Th contents were calculated from  $\gamma$ -ray peaks of their daughter nuclides ( $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  for U;  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  for Th), by assuming radioactive equilibrium throughout the U and Th series. The contents of U and Th are given by ppm and K by %, and  $^{137}\text{Cs}$  activity by pCi/g. The  $^{137}\text{Cs}$  activity was normalized to the sampling date.

The soil and sand are considered to be formed mainly by the weathering of bed-rocks. Therefore, the contents of natural U, Th and K reflect the composition of original rocks, though some fraction of these elements might have been depleted or enriched by the weathering processes and/or by the interaction of glacial meltwater. The following features can be deduced from the data given in Tables 2 and 3.

(1) The U, Th and K contents differ largely with the sampling sites, but the K/Th and U/Th ratios are not much different from each other (Fig. 3). The U, Th and K contents in the soils at Cape Royds are 3.3–4.3 ppm, 12.3–14.9 ppm and 2.5–3.7%, respectively, which are on an average 2 to 3 times higher than the soils in the Dry Valleys area, and just the same level as granite. On the other hand, the soil from

Table 2. The U, Th, K and fallout  $^{137}\text{Cs}$  contents in surface soils of the McMurdo Sound region, Southern Victoria Land, Antarctica.

Sampling location	Depth (cm)	U (ppm)	Th (ppm)	K (%)	$^{137}\text{Cs}$ (pCi/g)
Crater Hill (Ross Island)	0- 3	1.43±.05	8.5±.2	2.00±.04	0.019±.003
	3- 6	1.46±.06	8.1±.2	1.93±.06	n.d
	6-10	1.95±.05	8.8±.3	2.12±.06	n.d
Cape Royds (Ross Island)	0- 2	3.74±.07	12.3±.4	3.35±.08	0.18 ±.02
	2- 4	3.62±.03	12.9±.4	3.69±.09	0.02 ±.01
	4- 7	3.80±.07	12.8±.4	3.69±.07	n.d
	7-10	3.33±.12	12.8±.6	3.49±.07	n.d
	10-13	3.97±.11	13.7±.6	3.47±.09	n.d
Lake Vanda Delta area (Wright Valley)	0- 3	1.08±.05	7.1±.3	1.24±.05	n.d
	3- 6	1.15±.05	6.2±.3	1.07±.05	n.d
	6-10	0.99±.06	4.8±.2	1.02±.05	n.d
	10-15	0.88±.06	4.7±.2	1.04±.03	n.d
	15-20	0.57±.03	4.0±.2	1.09±.03	n.d
Morain sand near Canopus Pond (Wright Valley)	0- 3	0.93±.05	5.5±.3	1.43±.03	n.d
	3- 6	1.02±.05	7.5±.4	1.43±.03	n.d
	6-10	1.06±.04	5.6±.5	1.45±.05	n.d
	10-15	1.17±.04	6.4±.4	1.64±.04	n.d
	15-20	1.40±.07	7.6±.4	1.97±.04	n.d
Don Juan Pond (Wright Valley South Fork)	0- 1.5	0.62±.05	3.2±.2	0.94±.03	0.020±.004
	1.5- 3	0.70±.05	3.0±.2	0.89±.03	0.011±.004
	3- 5	0.66±.05	3.2±.2	0.90±.03	n.d
	5- 7.5	0.72±.05	3.2±.2	0.94±.03	n.d
	7.5-10	0.76±.05	3.6±.3	0.96±.03	n.d
Don Quixote Pond (Wright Valley North Fork)	0- 1	0.45±.05	2.1±.1	0.51±.03	0.034±.004
	1- 5	0.75±.05	3.0±.2	0.68±.03	n.d
	5-10	0.59±.04	2.3±.1	0.57±.03	n.d
Lake Fryxell (Taylor Valley)	0- 1	1.37±.04	5.4±.3	1.93±.09	0.56 ±.02
	1- 2	1.47±.04	6.9±.3	2.25±.03	0.04 ±.02
	2- 4	1.34±.03	6.1±.3	1.86±.03	n.d
	4- 6	1.34±.04	5.8±.4	1.75±.03	n.d
	6- 9	1.47±.05	5.8±.4	1.74±.03	n.d
[Surface sands]	0- 5				
Cape Royds, Ross Island		4.34±.09	14.9±.3	2.46±.01	0.06 ±.01
Lake Vida, Victorial Valley		0.82±.04	4.0±.2	1.25±.04	n.d
Lake Victoria, "		0.97±.04	6.2±.2	1.61±.04	n.d
Lake Vanda, Wright Valley		0.64±.04	3.9±.2	1.09±.05	n.d
Onyx River, "		0.69±.04	2.9±.2	0.91±.04	n.d
Lake Bull, "		0.80±.04	4.5±.2	0.97±.04	n.d
Lake Bull, (with evaporite)		1.74±.07	6.9±.2	1.68±.03	n.d
Lake Bonney-W, Taylor Valley		1.02±.04	5.6±.2	1.22±.04	n.d
" (with evaporite)		1.01±.04	4.3±.1	1.41±.03	n.d
" ( " )		0.94±.05	4.6±.2	1.26±.06	n.d
" ( " )		0.63±.02	4.0±.1	1.07±.02	n.d

n.d: not detected by non-destructive gamma-spectrometry.

Table 3.  $^{137}\text{Cs}$  and Pu isotopes of surface soils in the McMurdo Sound region.

Sampling Site	Depth (cm)	$^{137}\text{Cs}$ (pCi/g)	Pu* (pCi/kg)	$^{235}\text{Pu}/\text{Pu}^*$ (dpm/dpm)	$\text{Pu}^*/^{137}\text{Cs}$ (dpm/dpm)
Crater Hill	0 -3	$0.019 \pm .003$	$0.43 \pm .11$	—	$0.022 \pm .006$
	3 -6	LTD	$0.09 \pm .02$	—	—
Don Juan Pond	0 -1.5	$0.020 \pm .004$	$0.44 \pm .06$	—	$0.022 \pm .005$
	1.5-3	$0.011 \pm .004$	$0.18 \pm .06$	—	$0.016 \pm .008$
Lake Fryxell**	0 -1	$0.56 \pm .02$	$8.96 \pm .30$	$0.12 \pm .01$	$0.016 \pm .002$

\* Total activity of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ .

\*\* Lichen grown area.

LTD: Less than detectable.

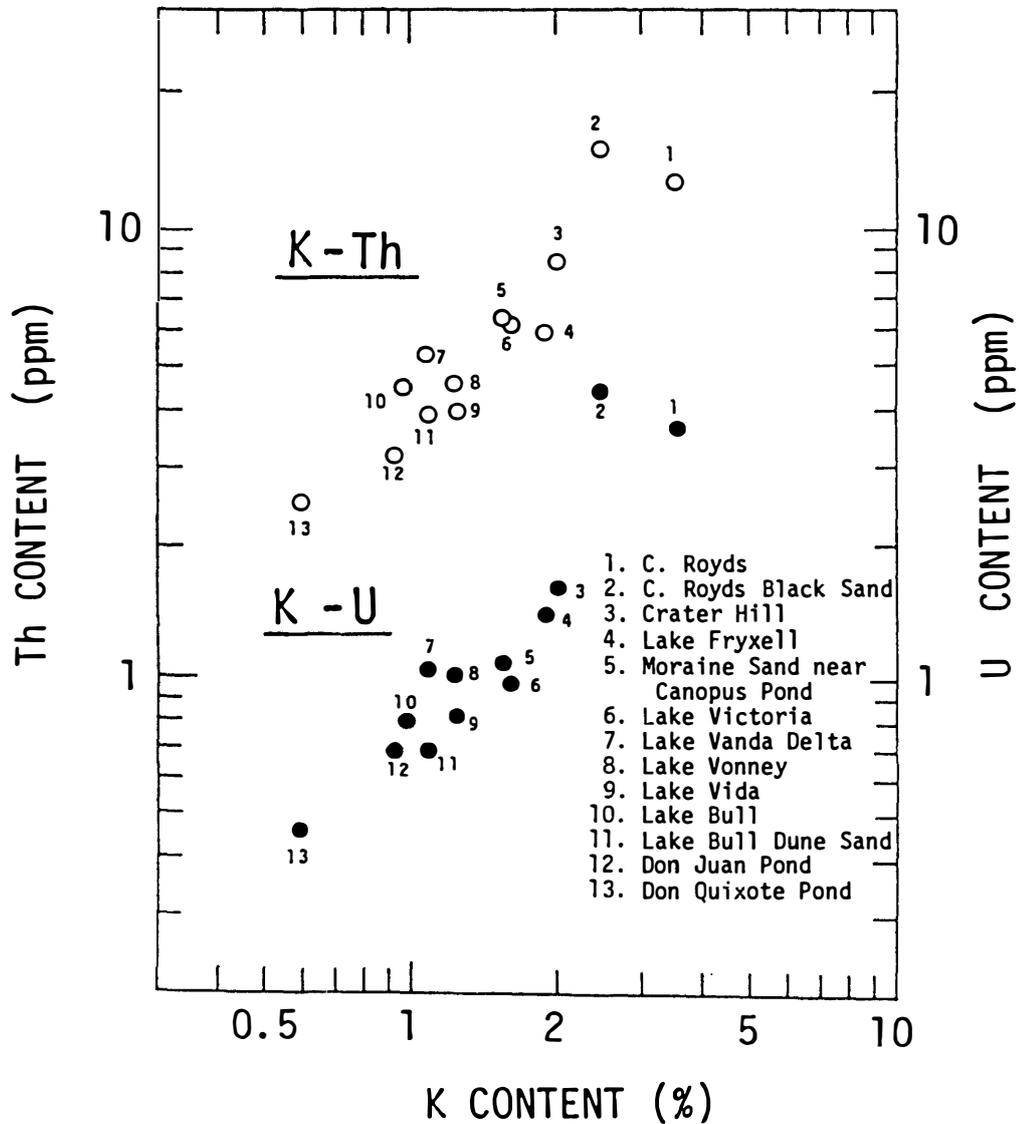


Fig. 3. Correlation of the K-Th and K-U contents for soils collected from the McMurdo Sound region, Antarctica. Open and closed circles show the correlations of K-Th and K-U, respectively.

Crater Hill behind the Scott Base showed not so high values as those of Cape Royds, though both soils are considered to have been formed by the weathering of the lava or the volcanic detritus from Mt. Erebus. The difference of the U, Th and K contents may indicate the different stages of the volcanic activity in Ross Island.

(2) The U, Th and K contents in the soils collected from the Don Juan Pond and the Don Quixote Pond are rather low. The soil sample of the Don Juan Pond was collected from the dried-up basin and that of the Don Quixote Pond was taken at the recently dried-up shore. Therefore, the low U, Th and K contents of these samples may be attributed to the dilution by the non-radioactive salt component abundantly existing in the pond water.

(3) The  $^{137}\text{Cs}$  activity is, in general, very low. This nuclide was detected only in the undisturbed surface soils within 4 cm in depth. Exceptionally high  $^{137}\text{Cs}$  content of 0.56 pCi/g was found for 0–1 cm layer of the soil collected from the lichen-grown area at the terminus of the Canada Glacier near Lake Fryxell, and this is explained by the accumulation of this nuclide in lichen tissue (HOLM and PERSSON, 1976). Accumulation of  $^{137}\text{Cs}$  per unit surface area was calculated from the  $^{137}\text{Cs}$  content and the sampling area to be about 1–5 mCi/km<sup>2</sup> (Table 4), which is one to two orders of magnitude lower than the values measured for the surface soils in Japan (YAMAMOTO *et al.*, 1983), indicating that the contamination due to artificial nuclides in Antarctica still remains at a low level as compared with other regions, particularly the temperate zone of the northern hemisphere (UNSCEAR, 1982).

Table 4.  $^{137}\text{Cs}$  accumulation in surface soil.

Location	$^{137}\text{Cs}$ accumulation (mCi/km <sup>2</sup> )
Cape Royds	5.5 ± 1.0
Crater Hill	0.9 ± 0.3
Don Juan Pond	1.0 ± 0.2
Don Quixote Pond	0.9 ± 0.2
Lake Fryxell	4.8 ± 0.2

(4) The  $^{239,240}\text{Pu}$  activity in surface soil was also found to be very low (Table 3). However, the  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio ranging from 0.016 to 0.022 lies just in the same range of the global fallout value. This indicates that the accumulation level of fallout nuclides precipitated on the ground surface in Antarctica can be well estimated by the measurement only of  $^{137}\text{Cs}$ . On the other hand, evidence of  $^{238}\text{Pu}$  release caused by the accidental burn-up of SNAP-9A satellite was clearly observed in the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratio (0.12 ± 0.01), which is about 3 times higher than that of the global fallout value of 0.03–0.04 measured for surface soils in Japan (YAMAMOTO *et al.*, 1983).

#### 4.2. Water filter

Nine water filters have been measured both by non-destructive  $\gamma$ - and  $\alpha$ -spectrometry. The results of measurements are summarized in Tables 5 and 6. Activities

Table 5. Radionuclide found in water filters used at Scott Base. 2-3 tons of snow meltwater was filtered by each filter.

Sample	F7911	F8001	F8002	F8003	F8004	F8005	F8006	F8007	F8008
Date of removal	30/11/79	?/1/80	18/2/80	28/3/80	6/5/80	11/7/80	?/?/80	?/?/80	?/?/80
Nuclide	Content of radionuclides (dpm/filter)								
[Natural radionuclide]									
<sup>210</sup> Pb	880±20	414±10	561±10	507±10	815±14	1193±18	483±7	252±7	231±6
<sup>7</sup> Be	4900±100	—	—	—	—	—	—	—	—
[Artificial radionuclide]									
<sup>54</sup> Mn	2.0±.3	6.9±1.0 (F8001+...+F8005)							
<sup>60</sup> Co	2.8±.4	6.9±.6 (F8001+...+F8005)							
<sup>106</sup> Ru	69±8	—	—	—	34±12	24±12	—	—	—
<sup>125</sup> Sb	47±3	10±1	16±2	10±3	14±3	29±3	8±2	5±1	4±1
<sup>134</sup> Cs	1.1±.2	—	—	—	—	—	—	—	—
<sup>137</sup> Cs	1060±50	389±6	337±6	369±6	726±9	464±2	187±3	127±3	116±2
<sup>144</sup> Ce	190±10	—	—	16±6	117±23	167±15	60±15	32±11	24±9
<sup>155</sup> Eu	15±2	5.1±.8	11±1	5.4±.8	12±1	16±1	6.6±.9	4.2±.7	3.3±.5
<sup>207</sup> Bi	2.5±.3	1.8±.6	2.4±.8	4.4±.4	5.9±.6	4.1±.5	1.7±.3	—	0.4±.2
<sup>239,240</sup> Pu*	16.6±1	11.4±.7	8.7±.6	8.6±.5	1.6±.4	13.5±.7	4.3±.3	2.4±.2	1.7±.2
<sup>241</sup> Am	5.0±.5	3.6±.5	2.8±.2	3.3±.2	5.2±.4	5.4±.4	1.8±.3	1.2±.2	1.0±.2

\* — Total activity of <sup>239</sup>Pu and <sup>240</sup>Pu.

Table 6. Activity ratios of radionuclides found in used water filters.

Filter sample	Activity ratio (dpm/dpm)				$^{230}\text{Th}/^{232}\text{Th}$
	$^{238}\text{Pu}/\text{Pu}^*$	$^{241}\text{Am}/\text{Pu}^*$	$^{137}\text{Cs}/\text{Pu}^*$	$^{234}\text{U}/^{238}\text{U}$	
F7911	$0.21 \pm .02$	$0.30 \pm .03$	$0.016 \pm .002$	n.m	n.m
F8001	$0.16 \pm .02$	$0.32 \pm .05$	$0.029 \pm .002$	$1.04 \pm .02$	$0.57 \pm .04$
F8002	$0.12 \pm .02$	$0.32 \pm .04$	$0.026 \pm .002$	$1.07 \pm .05$	n.m
F8003	$0.10 \pm .02$	$0.38 \pm .04$	$0.023 \pm .002$	$1.03 \pm .03$	$0.76 \pm .09$
F8004	$0.11 \pm .01$	$0.31 \pm .03$	$0.023 \pm .002$	$1.07 \pm .03$	n.m
F8005	$0.09 \pm .01$	$0.40 \pm .04$	$0.029 \pm .002$	$1.10 \pm .05$	n.m
F8006	$0.22 \pm .04$	$0.41 \pm .08$	$0.023 \pm .002$	$1.05 \pm .05$	n.m
F8007	$0.16 \pm .02$	$0.57 \pm .11$	$0.015 \pm .002$	$1.11 \pm .04$	n.m
F8008	$0.11 \pm .02$	$0.48 \pm .11$	$0.023 \pm .002$	$1.10 \pm .04$	n.m

\* Total activity of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ .

n.m: not measured.

are normalized to the date of the removal of each filter except for F8006, F8007 and F8008, and October 1st of 1980 was adopted for normalization. The contents of U, Th and K are not listed in Table 5, since these elements are considered to be originated from the air-borne dust of soil component and do not provide information of interest from the viewpoint of environmental radioactivity. Instead of the U and Th contents, activity ratios of  $^{234}\text{U}/^{238}\text{U}$  and  $^{230}\text{Th}/^{232}\text{Th}$  are shown in Table 6.

Figures 4 and 5 show the examples of Ge(Li) and Ge-LEPS spectra of water filter, respectively. Different from the case of surface soil, besides  $^{137}\text{Cs}$ , a number of artificial nuclides,  $^{60}\text{Co}$ ,  $^{106}\text{Rh}$  (supported by its long-lived precursor  $^{106}\text{Ru}$ ),  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{144}\text{Ce}$  and  $^{207}\text{Bi}$  were detected by the Ge(Li) measurement (Fig. 4), and  $^{210}\text{Pb}$  (natural fallout),  $^{155}\text{Eu}$  and  $^{241}\text{Am}$  could further be identified by Ge-LEPS measurement as seen in Fig. 5. Short-lived  $^7\text{Be}$  produced by cosmic-ray interaction with air component was detected in F7911 water filter sample measured soon after the removal

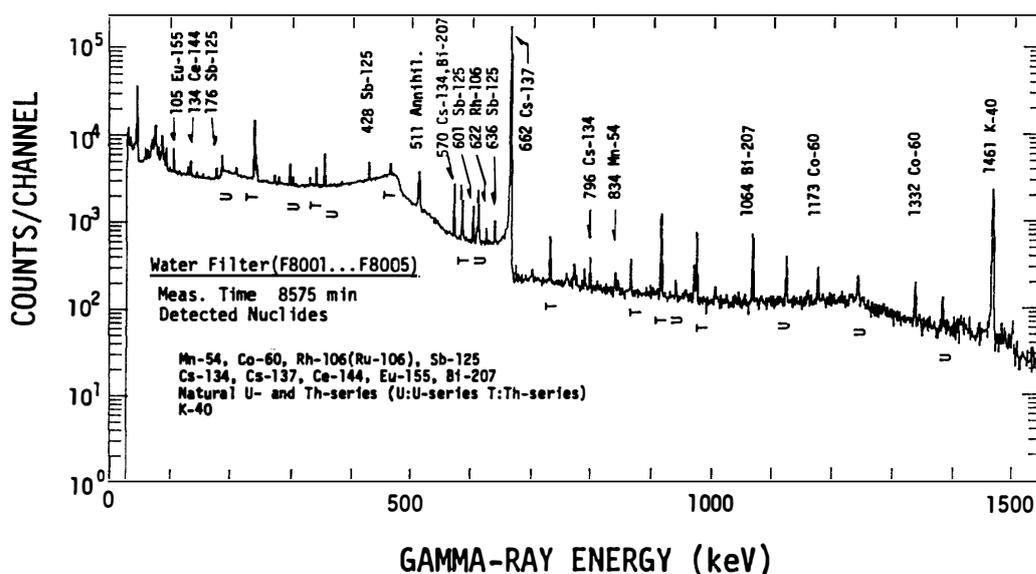


Fig. 4. Ge(Li) spectrum of water filter used at Scott Base. In order to detect low level radionuclides, combined sample of F8001 to F8005 was measured.

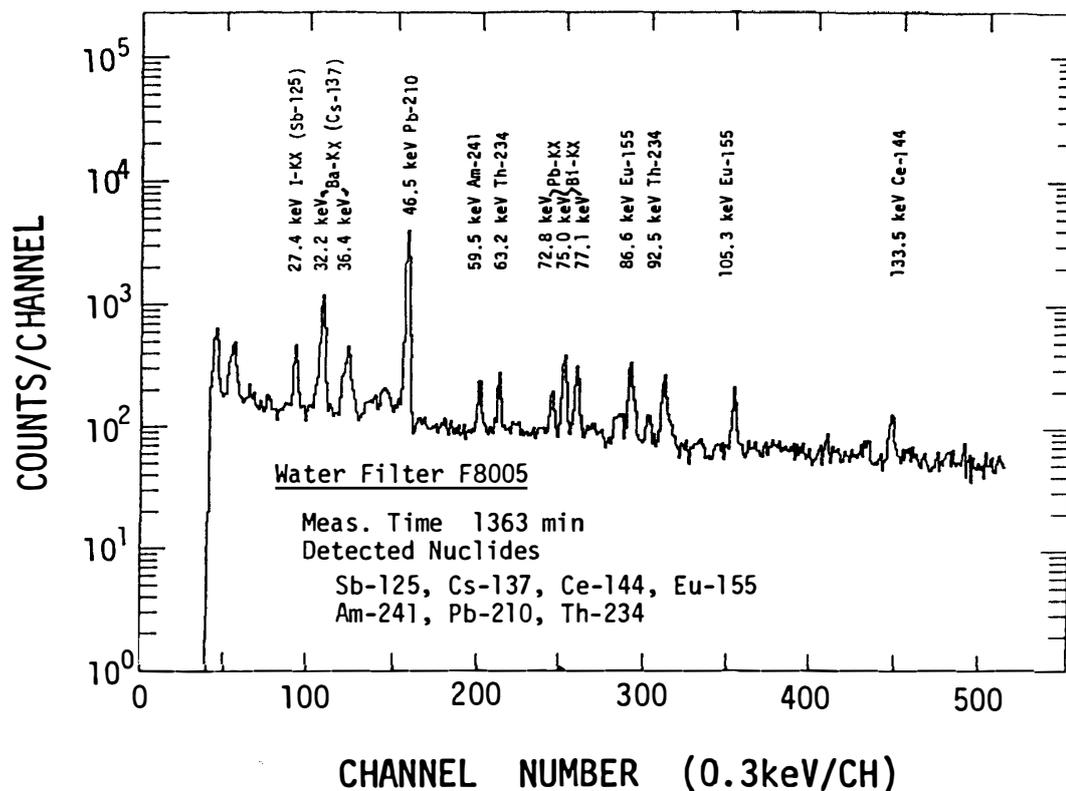


Fig. 5. Ge-LEPS spectrum of water filter sample F8005.

of the water filter.

The results of the measurements can be summarized as follows.

(1) The highest activity was found for artificial  $^{137}\text{Cs}$  and natural  $^{210}\text{Pb}$ . The former nuclide is produced by the nuclear explosion tests performed in the past three decades with high yield, and the latter is produced steadily by the decay of air-borne  $^{222}\text{Rn}$ . The accumulation level of both nuclides in Antarctica, however, is considered to be one to two orders of magnitude lower than that in other areas as discussed in the previous section. On the other hand, the accumulation level of other nuclides is much lower than that of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$ . A considerable portion of short-lived nuclides such as  $^{144}\text{Ce}$  and  $^{106}\text{Ru}$  is considered to have decayed, and low activities of  $^{54}\text{Mn}$ ,  $^{134}\text{Cs}$ ,  $^{125}\text{Sb}$ ,  $^{155}\text{Eu}$  and  $^{60}\text{Co}$  can be explained by both the decay and the low yield of these nuclides.

(2) Detection of  $^{207}\text{Bi}$  must be emphasized. This nuclide can hardly be detected in recently collected surface soils and fallouts. The only  $^{207}\text{Bi}$  data available are the measurements for marine organisms sampled near Eniwetok and Bikini atolls (WELANDER, 1969), where extensive nuclear explosion tests had been conducted in the 1954–1962 period. Recently we have detected  $^{207}\text{Bi}$  in the undisturbed surface soil collected from the forest in Toki City, Gifu Prefecture. The  $^{207}\text{Bi}/^{60}\text{Co}$  activity ratio was about 2, which is nearly the same as the one measured in the water filter samples. This suggests that  $^{207}\text{Bi}$  may possibly be detected in recent environmental samples by applying proper pre-concentration of the sample. Quite low  $^{207}\text{Bi}$  activity in recent environmental samples is probably due to the improvement of construc-

tion material of nuclear bomb and also to the decrease of nuclear test explosion in the atmosphere.

The detection of  $^{207}\text{Bi}$  in the Antarctic ice sheet indicates that the ice taken from the 'Cliff' contains the snow of at least 20–30 years old. The existence of recently accumulated snow is also suggested by the detection of short-lived  $^7\text{Be}$ .

(3) The contents of plutonium isotopes and  $^{241}\text{Am}$  are rather high as compared with  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  which are considered to have been produced by neutron-induced reaction on the construction material of nuclear bomb. The  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio is measured to be 0.015–0.029, which is in the same range as the one obtained from the surface soils shown above (Table 3) and the global fallout values. Any appreciable difference was not found also for the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio ranging from 0.30 to 0.57, which lies in the same range of the global fallout values (YAMAMOTO *et al.*, 1982, 1983).

(4) A large difference from the global fallout values was found for the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratios. The  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratios obtained from water filter samples range from 0.09 to 0.22, which are 3 to 5 times higher than the global fallout values measured for surface soils in Japan (YAMAMOTO *et al.*, 1983). According to the measurement by MIYAKE and SUGIMURA (1976), the increase of the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratio in surface sea water due to  $^{238}\text{Pu}$  of SNAP-9A was detected widely throughout the Pacific region, particularly in the high latitude areas of the southern hemisphere. As mentioned above, the high  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratio in the Antarctic samples can be attributed to the accidental release of  $^{238}\text{Pu}$  by the failure in the orbit of the SNAP-9A satellite which occurred in the southern hemisphere over the Indian Ocean in 1964.

(5) The activity ratios of natural uranium isotopes  $^{234}\text{U}/^{238}\text{U}$  range from 1.03 to 1.11, which is slightly higher than the equilibrium value. This may be explained by considering that the uranium in snow meltwater is composed of the uranium derived from wind-carried soil component having the  $^{234}\text{U}/^{238}\text{U}$  ratio of equilibrium value, the uranium derived from air-borne salt from sea water having the  $^{234}\text{U}/^{238}\text{U}$  ratio of 1.15 and the uranium supplied by glacier meltwater having rather high  $^{234}\text{U}/^{238}\text{U}$  ratio through the rock weathering.

(6) The  $^{230}\text{Th}/^{232}\text{Th}$  activity ratio was measured to be 0.57 and 0.76, which agrees well with the value of 0.5–0.8 estimated from the U and Th contents (Table 2) in surface soils by assuming radioactive equilibrium of both U and Th series nuclides. The origin of thorium isotopes found in water filters is considered to be the same as that of uranium, but the contribution of air-borne salt from sea water seems negligibly small because the content of thorium isotopes in sea water is several orders of magnitude lower than that of uranium isotopes. The main contributor of Th isotopes accumulated on snow may be attributed to the air-borne soil component from the surrounding area. The measurement of the  $^{228}\text{Th}/^{232}\text{Th}$  activity ratio is considered very useful to know the origin of thorium. However, since  $^{232}\text{U}$  equilibrated with  $^{228}\text{Th}$  was used as the yield tracer of uranium and thorium, the  $^{228}\text{Th}/^{232}\text{Th}$  ratio was not obtained in the present study.

The present data are considered to be useful for understanding the recent level of surface contamination of Antarctica. Measurement of the water filters recently sent to our laboratory is in progress and the results will be presented in the near future.

### Acknowledgments

The authors extend their sincere thanks to New Zealand Antarctic Research Programme, DSIR of New Zealand, and to Drs. Y. YUSA and S. NAKAYA for helpful guidance and assistance in the sampling of environmental samples. They also express their gratitude to Mr. I. KATO and the students of our laboratory for their assistance in radioactivity measurement.

### References

- CLAUSEN, H. B. (1973): Dating of polar ice by  $^{32}\text{Si}$ . *J. Glaciol.*, **12**(66), 411–416.
- CROZAZ, G. E., PICCIOTTO, E. and DE BREUCK, W. (1964): Antarctic snow chronology with  $\text{Pb}^{210}$ . *J. Geophys. Res.*, **69**, 2597–2604.
- FEELY, H. W., TOONKEL, L. E. and LARSEN, R. J. (1980): Radionuclides and trace metals in surface air. *Environ. Q.*, **EML-381**, Appendix-C, C1–C193.
- HOLM, E. and PERSSON, R. B. R. (1976): Transfer of fall-out plutonium in the food-chain lichen→reindeer→man. *Transuranium Nuclides in the Environment*. Vienna, IAEA, 435–446 (STI/PUB/410).
- LANGWAY, C. C., OESCHGER, H., ALDER, B. and RENEUD, A. (1965): Sampling polar ice for radiocarbon dating. *Nature*, **206**, 500–501.
- LOCKHART, L. B., PATTERSON, R. L. and SAUNDERS, A. W. (1965): Atmospheric radioactivity in Antarctica 1956–1963. *U. S. Naval Res. Lab. Rep.*, **6341**, 16 p.
- MARTELL, E. A. (1959): Atmospheric aspects of strontium-90 fallout. *Science*, **129**, 1197–1206.
- MIYAKE, Y. and SUGIMURA, Y. (1976): The plutonium content of Pacific Ocean waters. *Transuranium Nuclides in the Environment*. Vienna, IAEA, 91–105 (STI/PUB/410).
- OESCHGER, H., ALDER, B., LOOSLI, H., LANGWAY, C. C. and RENAUD, A. (1966): Radiocarbon dating of ice. *Earth Planet. Sci. Lett.*, **1**, 49–54.
- PICCIOTTO, E. and WILGAIN, S. (1963): Fission products in antarctic snow, a reference level for measuring accumulation. *J. Geophys. Res.*, **68**, 5965–5972.
- PICCIOTTO, E., CROZAZ, G. and DE BREUCK, W. (1964): Rate of accumulation of snow at the South Pole as determined by radioactive measurements. *Nature*, **203**, 393–394.
- UNSCEAR (1982): Exposure resulting from nuclear explosions. Ionization; Sources and Biological Effects. New York, United Nations, Annex E, 211–248.
- WELANDER, A. D. (1969): Distribution of radionuclides in the environment of Eniwetok and Bikini atolls, August 1964. *Symposium on Radioecology*, ed. by D. J. NELSON and F. C. EVANS. CONF-670503, 346–354.
- YAMAMOTO, M., KOMURA, K. and SAKANOUÉ, M. (1982): Distribution and characteristics of plutonium and americium in soil. *Environmental Migration of Long-Lived Radionuclides*. Vienna, IAEA, 481–489 (STI/PUB/597).
- YAMAMOTO, M., KOMURA, K. and SAKANOUÉ, M. (1983):  $^{241}\text{Am}$  and  $\text{Pu}$  in Japanese rice-field surface soils. *J. Radiat. Res.*, **24**, 238–249.

*(Received February 13, 1984; Revised manuscript received April 16, 1984)*