J. Radiat. Res., 47, Suppl., A85–A94 (2006)

Plutonium and Uranium in Human Bones from Areas surrounding the Semipalatinsk Nuclear Test Site

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Human bone/Plutonium/Uranium/Semipalatinsk nuclear test site/Internal dose.

To evaluate the present levels of ^{239,240}Pu and U in residents living near the Semipalatinsk nuclear test site, more than 70 bone samples were obtained at autopsy. The subjects ranged in age from 30 to 86 years (mean 59.3±12.9). Most of the samples consisted of victims who died of various diseases. Plutonium and U were radiochemically separated and determined by α-ray spectrometry. The mean concentrations of ^{239,240}Pu and ^{238}U observed were 0.050±0.041 mBq/g-ash (vertebrae 71, long-bones 18) and 0.28±0.13 mBq/g-ash (22.8±10.6 μg U/kg-ash) (vertebrae 58, long bones 16), respectively. The present ^{239,240}Pu levels were within the range found for human bone samples from other countries due solely to global fallout in the early 1980s. The average U concentration was close to the estimate (mean 22.5 μg U/kg-ash) for the UK, and about 10 times higher than those estimated for residents in New York City and Japan. By assuming that the average concentration of ^{239,240}Pu in bone samples is the value at 45 years after instantaneous inhalation in 1955, the initial total intake and the effective dose for 45 years were estimated as 10 Bq and 0.2 mSv, respectively. The annual intake of total U (^{234,235,238}U) and its effective dose for 60 years were estimated as 30 Bq for adult and 0.1 mSv, respectively, for chronic ingestion.

INTRODUCTION

Over a period of 40 years (1949–1989), the former USSR conducted more than 450 nuclear atomic explosions, including atmospheric, above-ground and underground nuclear tests at the proving ground (an area of about 18,000 km^2), 160 km west of Semipalatinsk City in Eastern Kazakhstan.[1,2] These experiments, especially atmospheric and above-ground tests, resulted in the global and localized dispersion of radioactive materials. Atmospheric tests finally stopped in 1962. Considerable efforts have been made to assess the radiation doses from nuclear tests to the general population living in areas near the Semipalatinsk nuclear test site (SNTS).[3–7] Since 1994, we have visited these areas several times to obtain baseline information on the radiological contamination. Long-lived radionuclides ^{137}Cs and ^{239,240}Pu on land at various areas have been measured and external radiation doses evaluated in several settlements by thermoluminescence (TL) using brick samples.[8–17]

Human tissue samples (mainly bone samples) were also obtained by autopsy from residents to estimate the internal exposure to people living near the SNTS. The concentration level and distribution in the human body of Pu and other actinide elements due to global fallout have been studied world-wide for the general population.[18–24] Around the SNTS, available data for α-emitters such as ^{239,240}Pu and U are, however, very sparse because of the difficulty of sampling and measurements. Human bone accumulates maximum amounts of Pu and U, and also retains them for a long time. Our earlier surveys around the SNTS showed that ^{239,240}Pu on land and ^{238}U in drinking water were in rather high concentrations. Plutonium is one of the most hazardous radionuclides and information about its distribution and body burden in the present population in this area is very important in evaluating current and future human risks.

The objective of this study is to establish the present ^{239,240}Pu and ^{238}U levels in the general population living near the SNTS and to estimate the associated alpha doses. For this purpose, we measured ^{239,240}Pu and ^{234,238}U in bone samples from more than 70 autopsies of residents from areas surrounding the SNTS. In this paper, we report the results of ^{239,240}Pu and U (^{238}U and ^{234}U) in human bone samples. Our own findings are compared with the values reported

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from other countries.

MATERIALS AND METHODS

Samples

Bone samples, mainly vertebral bone, were collected at autopsy from more than 70 residents who died in various settlements and in Semipalatinsk City near the SNTS, 1999–2003 (Fig. 1). The year of birth, age at death, sex and occupational data were obtained for each case. The subjects ranged in age from 30 to 86 years, most being 50–70 years (average 59.3; σ =12.9) (Fig. 2). Most died of chronic, debilitating diseases and cancer, without sudden death. The specimens were stored in formalin after dissection, and transported to our laboratory after most of the formalin of the samples was removed. They were then dried at 105°C and incinerated at 500°C by gradual increases in temperature in a muffle furnace for 5–8 days. The weights of bone ash were 5–20 g, corresponding to about 50–200 g wet weight.

Measurements of Pu and U

Plutonium and uranium were radiochemically separated and purified using a modification of the method described earlier by Singh *et al.*[25] Briefly, bone ash sample (5–15 g) was decomposed by HNO_3 with occasional additions of H_2O_2, after spiking with known amounts of ^{242}Pu and ^{232}U tracers. The residue obtained by evaporating to dryness was dissolved in 3M HCl. After adding $SnCl_2$ and HI while stirring without boiling, Pu and U were coprecipitated under a pH of about 2 with Ca present in bone as oxalate by adding an oxalic acid solution. The precipitate was dried and heated at 650°C in a muffle furnace overnight and dissolved in

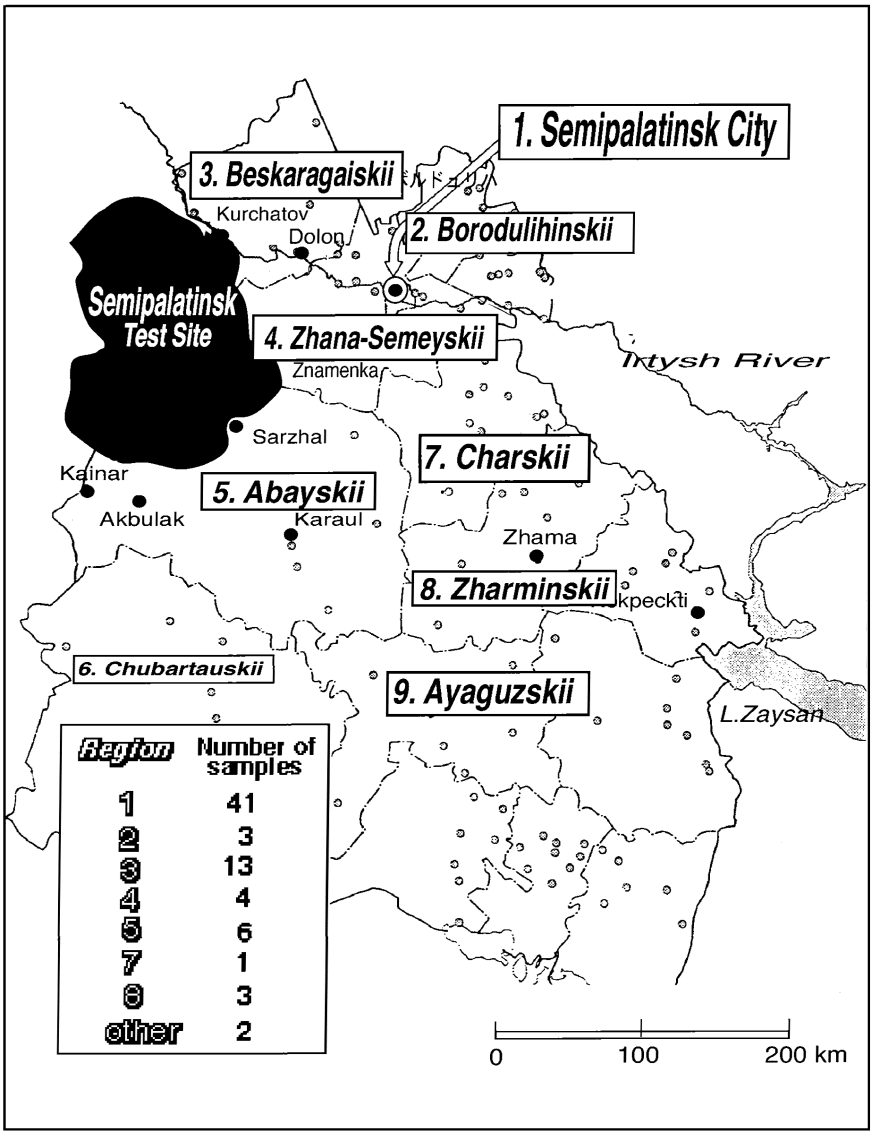

Fig. 1. Location map with bone sampling districts from residents around the Semipalatinsk nuclear test site (SNTS).

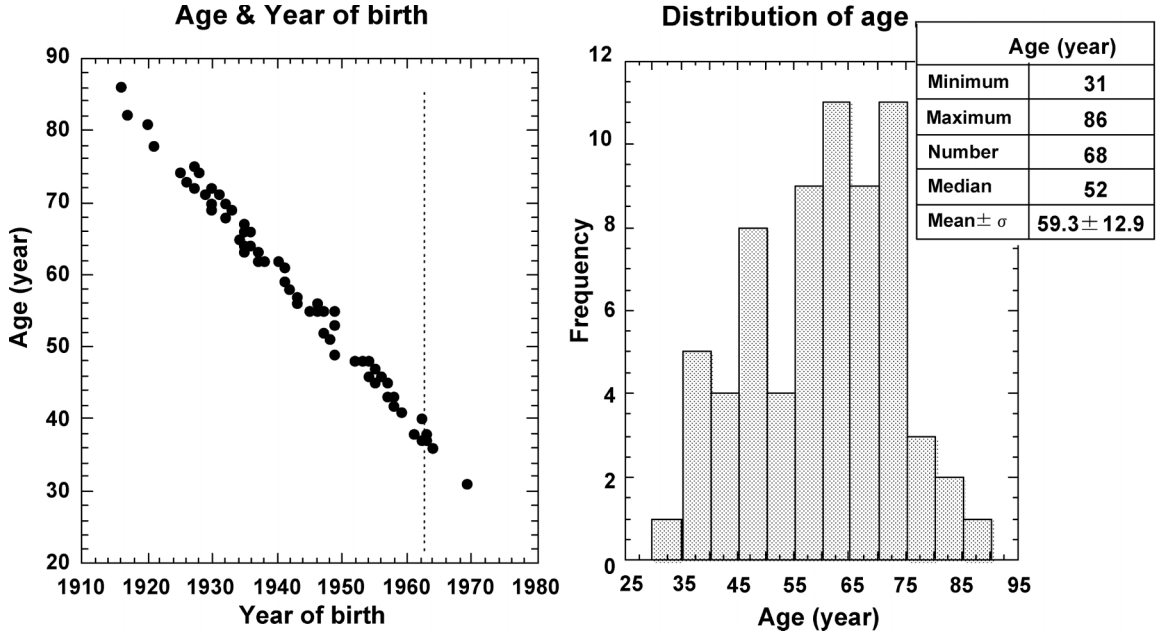

Fig. 2. Age at death vs. year of birth, and frequency distribution of age from residents around the SNTS. All samples were obtained from 1999 to 2003.

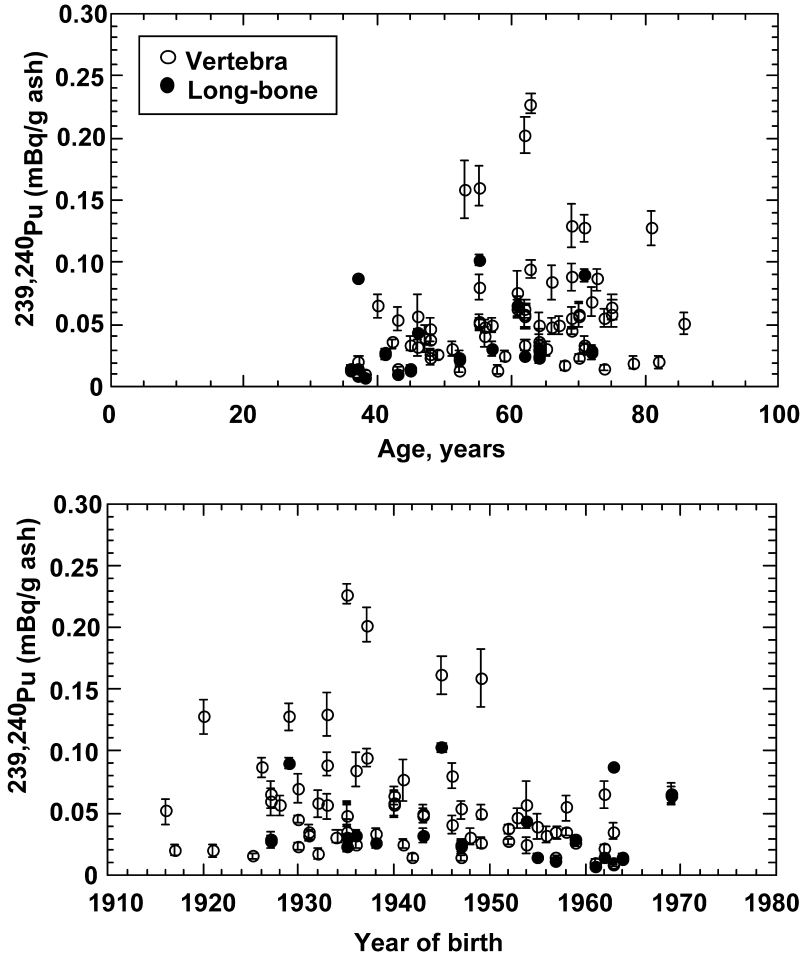

Fig. 3. Plots of ^{239,240}Pu concentrations in bone samples from residents around the SNTS vs. age at death or year of birth.

HNO_3 with heating. Then, Pu and U were coprecipitated with an iron carrier by adding ammonium hydroxide and the precipitate was dissolved in 8M HNO_3. Finally, Pu was purified by the usual anion exchange column method. The 8M HNO_3 fraction passing through the column containing most of the uranium was evaporated to dryness, and its residue was re-dissolved in 10M HCl. After removing most of the iron from the solution through solvent extraction with diisopropylether, the U was purified using the anion exchange column method as above. The chemical yields for both nuclides were more than 70%. The purified Pu and U were each electroplated onto a polished stainless steel disc and their activities were counted for more than one week by α-spectrometer using a surface barrier silicon detector and multichannel analyzers.

RESULTS AND DISCUSSION

Ideally all human tissue samples should be collected from victims of sudden death to minimize effects of disease processes on the liver or skeleton. Furthermore, large sample quantities should be analyzed because of non-uniformity of Pu within tissues. It is unfortunate that samples used here were taken from disease and cancer victims, and that sample amount was small. Nevertheless, it is important to evaluate the ^{239,240}Pu and U concentrations in human tissues at this point in time.

Plutonium in bone samples

The individual ^{239,240}Pu concentrations (mBq/g-ash) as a function of age and year of birth are shown in Fig. 3. The arithmetic mean, median and range of ^{239,240}Pu results from 89 bone samples (vertebrae 71, long bones 18) were 0.050±0.041, 0.036 and 0.007–0.23 mBq/g-ash, respectively (Fig. 4). By using the average ash-wet weight ratio of 10 % obtained from these samples, the corresponding ^{239,240}Pu concentrations in wet bone were 5.0±4.1, 3.6 and 0.7–23 mBq/kg-wet, respectively. The obtained ^{239,240}Pu levels were compared with previously reported values from several countries as shown in Table 1. It is evident that the present

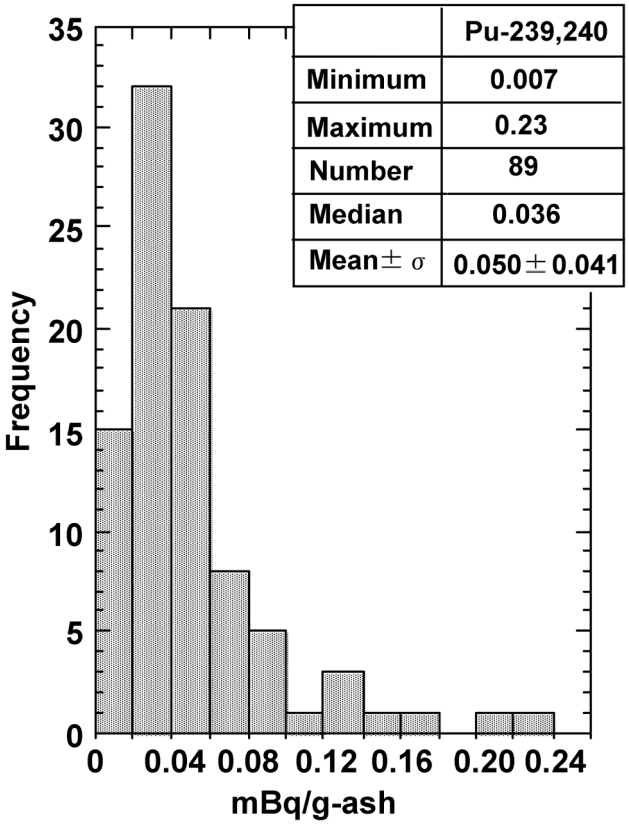

Fig. 4. Frequency distribution of ^{239,240}Pu concentrations in bone samples from residents around the SNTS.

Table 1. The concentration of ^{239,240}Pu (mBq/kg wet weight) in bone samples from non-occupationally exposed people.

Location	Year of Death	Vertebrae		Rib		Long bones		Reference
		mBq/kg wet	n	mBq/kg wet	n	mBq/kg wet	n	
Semipalatinsk area	1999–2003	5.4±4.3	(71)			3.7±2.9	(18)	This work
Tokyo	1978–1983	1.6±0.5**	(10)					(19)
Tokyo	1980–1982			8.5±5.9	(11) (rib+femur)			(20)
S. Finland*	1978–1979	3	(10)	2	(10)	2	(9)	(22)
New York City	1973–1974	9.3	(45)					(23)
Munich, F.R.G	1980–1981	3.7±1.8	(92)	2.9±1.3	(73)			(24)
Great Britain	1980–1984	5.1	(31)					(18)
Italy*	1982	10	(10)					(21)

*Median
*assuming 12.5% ash.

results are comparable to ^{239,240}Pu levels reported for bone samples from other countries in the early 1980s.

As previously reported, the current radiological situation near the SNTS is characterized by local fallout contamination.[9–17] The ^{239,240}Pu inventories are clearly elevated compared with the levels of global fallout ^{239,240}Pu found at this geographical latitude. For example, the values of several 100-several 1,000 Bq/m^2 have been found, with the highest values of over 10,000 Bq/m^2 in the village of Dolon which is located close to the plume axis of the first test explosion in August 1949.[15] Furthermore, in the southern areas around the SNTS, the ^{239,240}Pu inventories were found in the wide range of 34–2,050 Bq/m^2, most of the data being 100–300 Bq/m^2. Higher values of more than 1,000 Bq/m^2 were also observed in the areas surrounding the Karaul, Tailan and Sarzhal settlements through which the radioactive cloud related to the first thermonuclear explosion passed in August 1953.[16] However, although these areas are contaminated with elevated ^{239,240}Pu levels, the ^{239,240}Pu activity in locally-grown foodstuffs collected in or close to the village of Dolon is reported to be below the limit of detection.[26] A significant amount of ^{239,240}Pu in soil from these areas is associated with fused silicate. The fact that ^{239,240}Pu is not detected in the locally-grown food is generally explained by the low bioavailability of Pu around here, that is, the low root uptake and low transport along various food-chains of Pu.[26] Therefore, intakes of ^{239,240}Pu via ingestion from diet seem to be very low. Data on intake of ^{239,240}Pu via inhalation are very sparse. Measurements of levels in air taken at Dolon village over 1.5 years were reported to be as high as 0.08 mBq/m^3. Steinhäusler *et al*[27] estimate that this level (0.08 mBq/m^3) results in an annual effective inhalation dose of 0.03 mSv. So, contribution of ^{239,240}Pu intake from inhalation seems to be small, although we are now measuring the ^{239,240}Pu levels in human lung samples in this regard. Taking into account the very low intake of ^{239,240}Pu via ingestion and inhalation, the present ^{239,240}Pu levels found in the bone samples appear reasonable.

The age trend for ^{239,240}Pu concentration is shown in Table 2. The Pu levels for age groups of 51–60, 61–70 and 71–86 y are comparatively higher than the values in the 30–50 y age groups . When a two tailed Student's t-test (95% significance level, $\alpha = 0.05$) was attempted, significant differences among the age groups of 31–40 y, 41–50 y and 3 groups over 51 y were found. No significant difference was found in ^{239,240}Pu concentration among districts of people whose bone samples were obtained (two tailed Student's t-test; 95% significance level, $\alpha = 0.05$): 0.058 ±0.048 mBq/g-ash (n=38) in Semipalatinsk City and 0.044±0.034 mBq/g-ash (n=51) in other districts close to the SNTS such as Beskaragai, Abayskii, etc.

Table 2. Age dependency of ^{239,240}Pu concentration in bone samples (vertebrae: 58, long bones: 16) from the SNTS

Age range (y)	No. of samples	^{238}U activity (mBq/g-ash)
30–40	9	0.019±0.018
41–50	18	0.032±0.013
51–60	16	0.057±0.046
61–70	30	0.061±0.048
71–86	16	0.057±0.036

Uranium in bone samples

Uranium is a naturally occurring radioactive and heavy metal found in the earth. Natural uranium contains three isotopes, namely ^{238}U(99.27% by mass), ^{235}U(0.72%) and ^{234}U(0.0054%). ^{238}U and ^{235}U are the origin of two different decay chains. In nature, ^{238}U in rock and soil is usually in secular equilibrium with the daughter ^{234}U, that is, the activity ratio of ^{234}U/^{238}U is unity (i.e. 1.0). Uranium enters the human body via ingestion from drinking water and food, and inhalation. We have measured U isotopes in soil and drinking water samples from various areas near the SNTS, but anomalous ^{235}U/^{238}U ratios were not found, indicating that the source of U intake is of natural origin.[28]

The analytical results of the human bone samples (vertebrae 58, long-bones 16) are shown in Fig. 5 as a function of age of death and year of birth, as was done in the case of ^{239,240}Pu. Most of the samples (mean age: 57.9±12.8 y, range: 29–86 y) are the same as those used in the Pu analyses. The ^{238}U concentrations were found in the range 0.09 to 0.69 mBq/g-ash, within a factor of 10 (Fig. 6). These values correspond to 7.3–56 μg U/kg-ash in terms of U concentration. The mean and median values (n=74) were 0.28±0.13 and 0.25 mBq/g-ash (or 22.8±10.6 and 20.4 μg U/kg-ash), respectively. Measurements of U in different bone types do not reveal any marked differences in concentration among vertebrae (mean: 0.29±0.14 mBq/g-ash) and long-bones (mean: 0.23±0.11mBq/g-ash). A marked disequilibrium (mean 1.62±0.34, range 1.02–2.81) higher than the value of 1.0 was observed for ^{234}U/^{238}U activity ratios, as shown in Fig. 6. This is consistent with values from drinking water samples from various areas near the SNTS.

There have been several reports on uranium in human tissues of non-occupationally exposed humans. By analyzing 63 bone and cartilage samples in the UK, Hamilton *et al*[29] found an average concentration of 22.5 μg U/kg-ash (3.0 μg U/kg-wet, assuming 13.5% ash). For 58 vertebrae from New York City residents, Fisenne[30] observed a mean (median) value of 0.43±0.26 (0.32) μg U/kg-wet weight, whose values correspond to 3.2±1.9 (2.4) μg U/kg-ash. Ikeda *et al*[31] and Igarashi *et al*[32] reported mean values of 8.4 μg U/kg-ash (1.1 μg U/kg-wet) and 2.5 μg U/kg-ash (0.69 μg U/kg-wet), respectively, based on the measurement of bone samples from 5 Japanese subjects and 20 Japanese skull, rib and

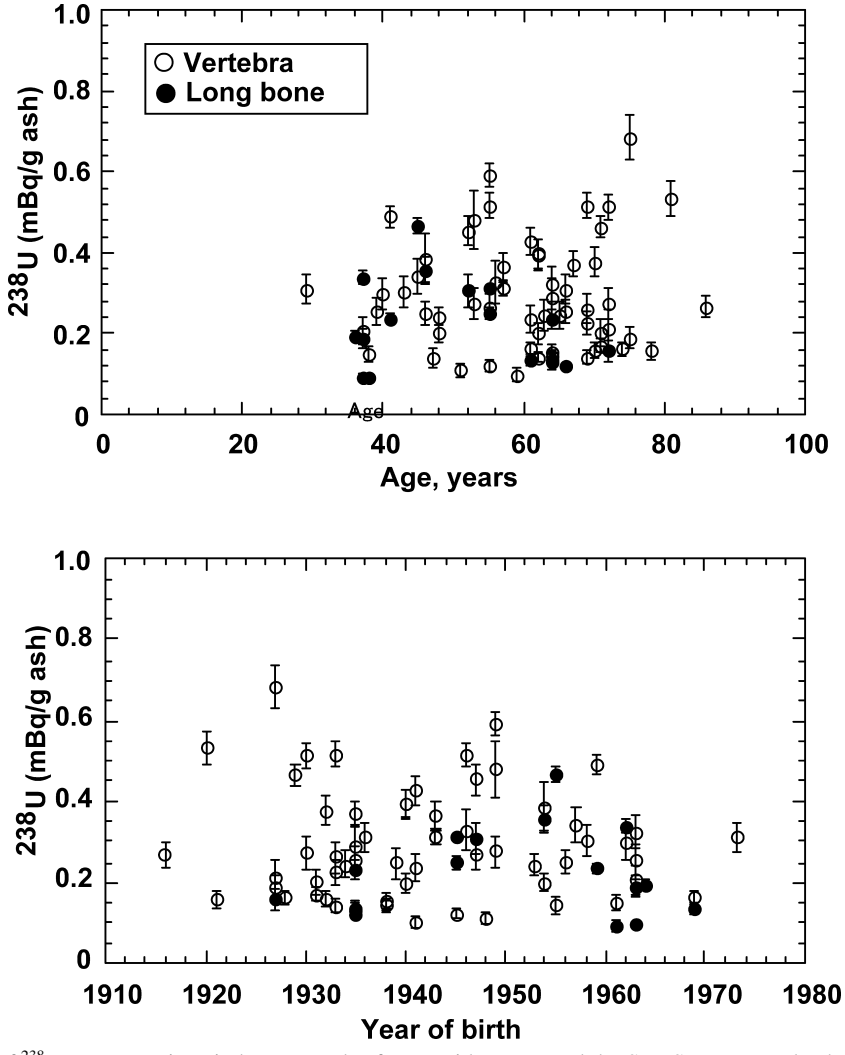

Fig. 5. Plots of ^{238}U concentrations in bone samples from residents around the SNTS vs. age at death or year of birth.

femur samples. The average U concentration found in the Semipalatinsk region is rather close to the estimate for the UK, and about 10 times higher than that estimated for New York City and Japanese residents. Thus, although there have been several reports on U in human bone samples, some apparent differences exist in these data. Such differences are considered to be due to geographical differences in uptake of this element. The normal dietary levels of U for people in Semipalatinsk area are unknown. The U concentrations in the daily diets of the UK, New York City and Japan vary by only a factor of 1.5 (1, 1.3 and 1.5 μg U/d, respectively).[30] Besides the dietary intake of U from food, the contents of U in drinking water from 5 Japanese cities and in New York City water are reported to be, respectively, 0.009 μg U/L on average and in the range from 0.03–0.08 μg U/L.[30,33] In England and Scotland, a somewhat high value of <1 μg U/L is observed.[29] We collected drinking water samples from some settlements in areas surrounding the SNTS and measured U concentrations (μg U/L): 3.8–26.9 (n=7) in Dolon, 1.9–3.4 (n=4) in Cheremushka, 9.3–12.2 (n=3) in Budene, 10.4–16.8 (n=4) in Sarzhal, 28.9 (n=1) in Karaul, 0.3–0.4 (n=2) in Kainar and <1 (n=3) in Semipalatinsk City. Most of these data found are several 10 to 100 times higher than those of the above mentioned countries, except for the U concentrations in Semipalatinsk City and Kainar village which are nearly the same as those reported for England. For the drinking water samples from the settlements, high ^{234}U/^{238}U activity ratios ranging from 1.5–7.9 (mostly 1.5–3.0) were also found. Such high U concentration in drinking water is likely to be responsible for the high U concentration in bone of people living near the SNTS. However, the differences in U concentration between districts of people whose bone samples were obtained were not shown to be significant at this time: 0.27±0.12 mBq/g-ash (n=35) in Semipalatinsk City and 0.29±0.14 mBq/g-ash (n=39) in other districts close to the SNTS such as Beskaragai, Abayskii, etc. The daily U intakes from foodstuffs must be further considered, together with the measurements of increasing num-

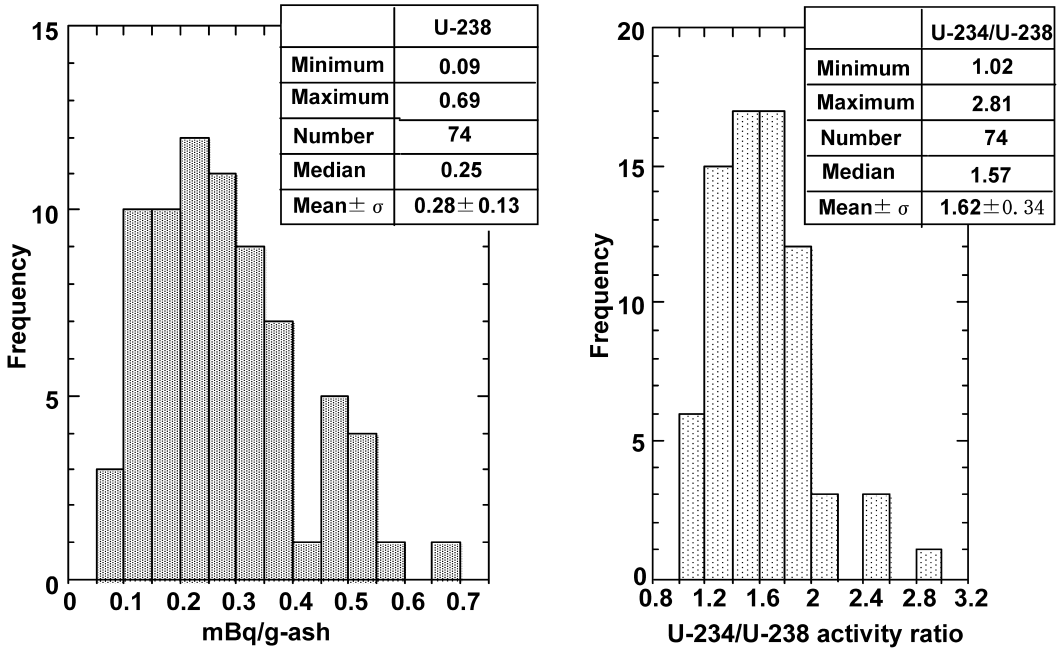

Fig. 6. Frequency distribution of ^{238}U concentrations in bone samples from residents around the SNTS.

bers of bone samples from the latter areas.

The measured U concentrations were separated into 5 age groups (30–40, 41–50, 51–60, 61–70 and 71–86 y of age) as shown in Table 3 and a two tailed Student's t-test (95% significance level, $\alpha = 0.05$) was examined for differences in ^{238}U concentration with age. A statistically significant difference with age was found for age groups between (30–40 y or 61–70 y) and 51–60 y. There is a report that the values obtained from New York City residents indicate an increasing U concentration in vertebrae with age.[30] On the other hand, no statistically significant difference is found for autopsy cases in Japan [33] or for autopsy cases in the UK.[29] Further studies are needed using samples from the victims of sudden death from the areas surrounding the SNTS.

Table 3. Age dependency of ^{238}U concentration in bone samples (vertebrae: 58, long bones: 16) from the SNTS.

Age range (y)	No of samples	^{238}U activity (mBq/g-ash)	U content* (μg/kg-ash)
30–40	9	0.23±0.08	18.7± 6.5
41–50	13	0.28±0.13	22.8±10.6
51–60	13	0.35±0.13	28.5±10.6
61–70	26	0.24±0.11	19.5± 9.0
71–86	13	0.31±0.18	25.3±14.7

*Calculated by using ^{238}U abundance of 99.27%.

Effective dose estimation for ^{239,240}Pu and U

For estimating radiation dose to residents near the SNTS, dose assessment requires specific information. The fundamental data to evaluate the radiation dose include concentrations of radionuclides in air and foods at the residential area under study, chemical and physical properties of radionuclides, and physiological characteristics of the residents. Because of the lack of them, a preliminary dose assessment was done with some assumptions. The average age at death of sample residents was about 60-years old and the average year of birth was in the early 1940s, so a reference resident was considered to be born in 1940 and to die at 60 y. For estimating the dose from Pu, it was assumed that inhalation was instantaneous in the middle year (1955) of the period of atmospheric tests; the inhalation solubility class was type S,[34] and the size of an aerosol particle was 1 μm in diameter. On the other hand, uranium was assumed to enter the human body via chronic ingestion from drinking water and food from birth to death, because U was considered to be natural in origin. The annual food consumption for an infant (1-year old) and a child (10-years old) was set at about 40% and 70%, respectively, of the adult value, based on the UNSCEAR 2000 report. [35]

The ICRP respiratory tract model[34] and the ICRP biokinetic models[36,37] were applied to evaluate intake, distribution and retention of radionuclide in the human organs and tissues. In the present work, the measured concentrations of ^{239,240}Pu and ^{238}U(and ^{234}U) in bone samples were defined as the average concentration in total mass of cortical and trabecular bone and bone marrow. Estimated burdens in lung, bone, liver and kidneys of ^{239,240}Pu for an instantaneous inhalation of unit activity in 1955 are shown in Fig. 7, together with those in bone, kidneys and liver of ^{238}U for chronic

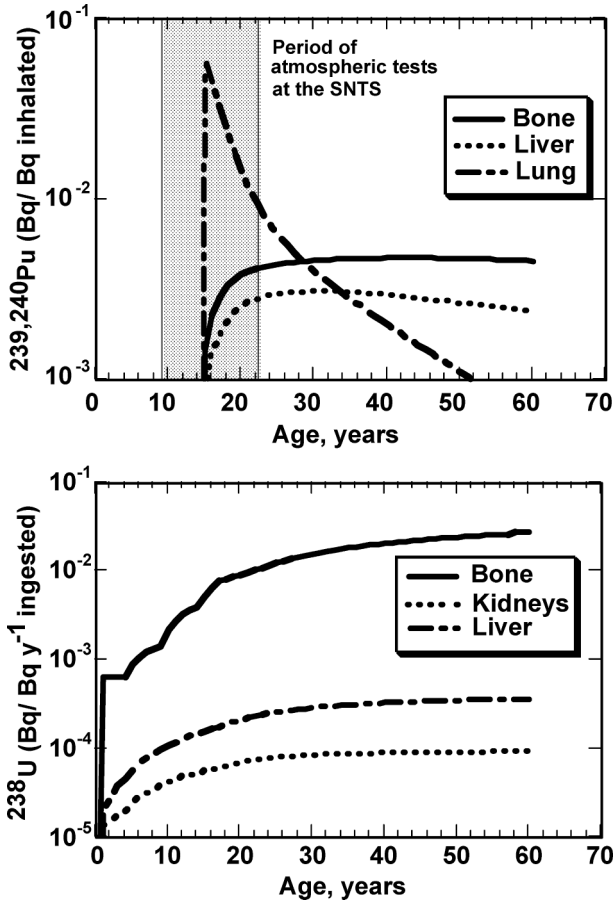

Fig. 7. Estimated burdens in lung, bone, liver ad kidneys of ^{239,240}Pu for an instantaneous inhalation of unit activity (1 Bq) in 1955, and ^{238}U for chronic ingestion at adult's rate of 1Bq/y. The ICRP 66 respiratory model and ICPR's 67 and 69 biokinetic models were applied to evaluate intake, distribution and retention of radionuclides in the human organs and tissues.

ingestion at the adult's rate of 1 Bq/y. If the measured average concentration of $^{239,\ 240}$Pu in bone samples was the value at 45-years after intake in 1955, the initial total intake and the effective dose for 45-years are estimated as 10 Bq and 0.2 mSv, respectively. Annual intake of ^{238}U and the effective dose for 60-years are also estimated as 10 Bq for adult and 0.03 mSv, respectively. If the activity ratios 1.62 of ^{234}U/ ^{238}U and 0.047 of ^{235}U/ ^{238}U in bone samples are the same as ratios in foods, the annual intake of total U and the effective dose for 60-years are calculated to be 30 Bq for adult and 0.1 mSv, respectively.

CONCLUSION

The ^{239,240}Pu and ^{234,238}U in bone samples of more than 70 autopsy cases of residents from areas surrounding the SNTS were determined to establish the present ^{239,240}Pu and ^{238}U levels in the general population living near the SNTS and to estimate the associated alpha doses for this population. The results are summarized as follows:

(1) The mean concentrations of ^{239,240}Pu were 0.050±0.041 mBq/g-ash (vertebrae 71, long-bones 18). The present ^{239,240}Pu levels were within the values reported from other countries for the bone samples due to global fallout in the early 1980s.

(2) The mean concentration of ^{238}U was 0.28±0.13 mBq/g-ash or 22.8±10.6 μg U/kg-ash (vertebrae 58, long bones 16). The average U concentration was close to the estimate (mean 22.5 μg U/kg-ash) for the UK, and about 10 times higher than those estimated for New York City and Japanese residents. A marked disequilibrium (mean 1.62±0.34, range 1.02–2.81) higher than the value of 1.0 was found for ^{234}U/^{238}U activity ratios. Such values are known for drinking waters from various areas near the SNTS.

(3) By assuming that the average concentration of ^{239,240}Pu in bone samples is the value at 45-years after instantaneous inhalation in 1955, the initial total intake and the effective dose for 45-years were preliminarily estimated as 10 Bq and 0.2 mSv, respectively.

(4) The annual intakes of total U(^{234,235,238}U) and the effective dose for 60-years were also estimated as 30 Bq for adult and 0.1 mSv, respectively, for chronic ingestion.

AKNOWLEDGEMENTS

The authors would like to express their gratitude to research staff of the Kazakh Scientific Research Institute of Radiation Medicine and Ecology for help with sampling. We are also grateful to Mr. M. Ohnishi and Mr. T. Miki of Fukui Prefectural Institute of Environmental Radiation Research and Monitoring Center for preparing the bone ash samples. This work was supported by a Grant-in Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture, Japan, during 1998–2003 (M. H. & M. Y.)

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Received on July 1, 2005
1st Revision received on July 1, 2005
Accepted on November 2, 2005