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Baseline quantity of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in urinary excretions from Thai people and internal exposure dose

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Abstract: Today, sealed and unsealed radioactive materials have been used in Thailand for various purposes such as medical, agricultural and industrial applications. There is a growing trend in the use of radioactive materials. Moreover, neighboring countries are planning to construct and operate nuclear power plants. In case of nuclear power plant accidents, radioactive releases in environment and intakes into human body by inhalation and ingestion causing long term health effects. This research aims to determine the radiation baseline quantity of interested relevant radionuclides such as ^{131}I, ^{137}Cs, ^{134}Cs as well as a natural radionuclide, ^{40}K in urine samples of Thai people by gamma spectrometry. Two types of detectors (NaI and HpGe detectors) are calibrated by mixed radionuclide standards of ^{109}Cd, ^{57}Co, ^{133}Ba, ^{54}Mn, ^{137}Cs and ^{60}Co, (energy range from 88 to 1,331 keV). 720 urine samples are collected over a 24 hour period from Thai volunteers with the age older than 18 years old, who lived in eight locations of Thailand. To reduce the effect of geometric difference, 30 ml of urine samples are prepared for counting measurement and efficiency determination. The radiation baseline quantity of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in 30 ml of urine samples are 0.37±0.09, 0.63±0.13, 0.39±0.08 and 7.84±1.63 Bq, respectively. Based on the assumption of intake (50% of the intake by ingestion and 50% of the intake by inhalation), internal dose for members of public are assessed. The committed dose equivalent due to an intake of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are 2.36E-03±1.66-03, 1.15E-01±8.61E-02, 1.16E-01±7.77E-02, 9.44E-01±3.56E-01 mSv per year, respectively.

1. Introduction

Radiological contamination has been widely found in the normal environment in Thailand, for example in the air, soil, food and drinking water. According to the wide spreading contaminants, radioactive material can enter through the body via ingestion and inhalation. Some radionuclides such as K-40 has been found as the primary source of radiation inside human body. Due to radioactive decay, the radiological contaminants may affect not only people but also plants and animals. The internal intake of radioactive material has been considered as one of the major concerns for radiation protection. To make sure that the level of the internal contamination is not higher than the accepted level; biological samples such as urine, excreted from human body should be monitored. Natural intake of radioactive material is determined and assessed.

This research mainly determined the quantity of radioactivity and assessed the internal contamination of radioactive material in urine samples, collected from volunteers of 8 locations in Thailand. The interested radionuclides include radioactive materials; leaked from nuclear accidents,

nuclear activities, nuclear fuel cycles and other applications; for example ^{134}Cs, ^{137}Cs, ^{131}I, ^{14}C, ^{90}Sr, ^{3}H, ^{241}Pu, ^{232}Th and ^{238}U. The radiation baseline of interested nuclide will be applied as reference data for Thai people in case of nuclear accidents both of Thailand and neighboring countries.

2. Methods

2.1. Network Creations

Collecting urine samples is difficult to manage by only one institute. Convenience sampling method is applied by network creations in order to provide volunteers who are conveniently available to participate in this research for collecting urine samples. The appropriated universities should have a gamma ray spectrometer or locate in suitable location where dispersed in many parts of Thailand. They were invited to participate as networks. Coordinated network with 10 universities where located in 10 parts of Thailand was created. Participated universities played a part in collecting and/or measuring radioactivity in urine samples. The urine samples and radioactivity determinations were submitted to the Office of Atoms for Peace (OAP) for the measurement of internal dose assessment. Unfortunately, the raw data of 2 locations had problems and could not be conclusion. The urine samples collecting will be repeated and reanalyzed next time. Therefore, there are only the results of 8 locations in this research as show in Figure 1.

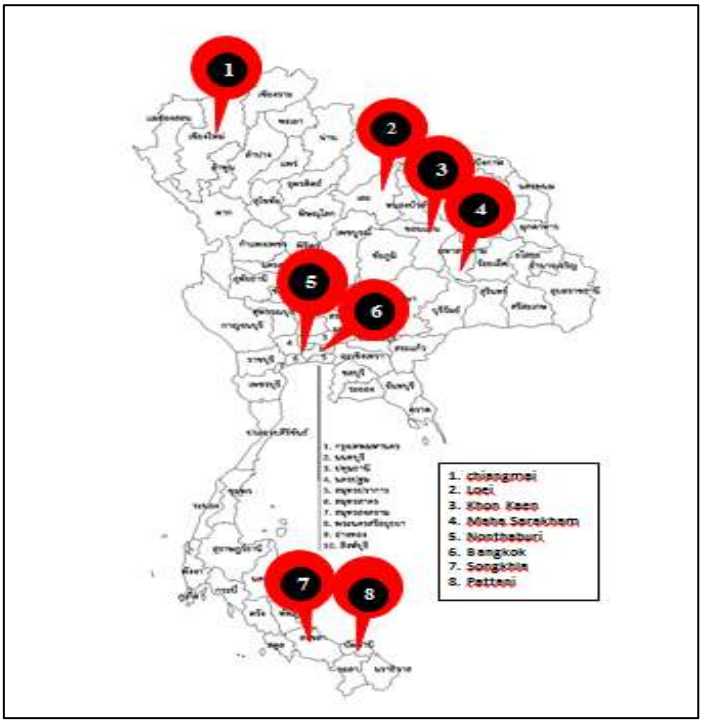

Figure 1. Urine sampling locations.

2.2 Urine sampling

Urine samples are collected over a 24 hour period from Thai volunteers with the age older than 18 years old, who lived in 8 locations of networks. The networks provided at least thirty volunteers for collecting urine samples which are kept in 500 ml of wide mouth polyethylene bottles. Urine samples are continuously collected for 3 days. There were at least 90 urine samples per each location. The total samples are 720.

2.3. Sample preparation

Urine samples are continuously collected for 3 days. Then, they were mixed together and the total volume was measured. 720 urine samples were prepared by adding 10% (v/v) of concentrated nitric acid. 30 ml of each sample was withdrawn, transferred to plastic bottle and measured by appropriated detector in order to determine the radiation baseline quantities of ^{131}I, ^{134}Cs, ^{137}Cs and ^{40}K. The rest of urine samples were kept in cool temperature for the next measurement.

2.4. Calibration

Two types of radiation detectors were used. NaI and HpGe detectors with Genie 2000 gamma spectroscopy software in this study were similarly calibrated by mixed radionuclide standards (point source) of ^{109}Cd, ^{57}Co, ^{133}Ba, ^{54}Mn, ^{137}Cs and ^{60}Co (energy range from 88 to 1,331 keV). The efficiency of each radionuclide was calculated in equation 1, when cps is counts per second, dps is activity in Bq and I is gamma ray abundance. The efficiencies from calibration were fitted a curve. Due to the difference of geometry between sample and calibration point source, the prepared solution containing ^{131}I in the same geometry type as the urine sample was used for correction counting efficiencies. The efficiency ratio(R) was determined by dividing the efficiency of ^{131}I in urine by the efficiency of point sources at the same energy. The efficiencies for urine sample counting were determined by using R value multiply by the efficiencies from calibration fitted curve of point sources.

$$\text{eff} = \frac{\text{cps}}{\text{dps x I}} \tag{1}$$

2.5. Gamma analysis

Referred to RS-G-1.2 [3], gamma emission from radionuclides in urine samples can be detected by NaI or HpGe detector with Genie 2000 spectroscopy software. To determine the quantities of radioactivity in urine samples, 30 ml of empty plastic bottle was used as a blank. Radioactivities of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K were measured at 356, 662, 796 and 1,461 keV respectively.

$$A = \frac{\text{cps}}{\text{eff}} \tag{2}$$

When eff is efficiency for urine sample counting of radionuclide multiplied by its gamma ray abundance.

2.6. Internal dose assessment

According to ICRP 78 [1], biokinetic models which describe activity in excreta, as a function of time following intake. To calculate an estimate of intake of radionuclides by the members of public, the daily activities of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in urine samples, M, is divided by the predicted values of the measured quantity in urine at time t days after intake of 1 Bq or m(t) as show in equation 3. Most of m(t) parameters were taken from ICRP 78 except for m(t) parameter of ^{40}K for ingestion was taken from Turban *et al.*, [4] and the inhalation was taken from calculation on CINDY code program.

$$\text{Intake} = \frac{\text{M}}{\text{m(t)}} \tag{3}$$

The intake can be multiplied by the effective dose coefficients, e(70), to give the committed effective dose equivalent(CEDE) as show in equation 4. The effective dose coefficients, e(70), were taken from Health Physics 2012 [2].

$$\text{CEDE} = \text{Intake} \times \text{e(70)} \tag{4}$$

2.7. Minimum detectable activity

The minimum significant activity (MSA) is related to the smallest recorded signal. It is a significant excess of the background response indicating the presence of radioactivity at 95% of probability which can be calculated in equation 5.. To ensure that the radionuclide signals correspond to the MSA, the minimum detectable activity (MDA) was calculated as equation 6.

$$\mathrm{MSA} = \frac{1.64}{\mathrm{F}} \sqrt{\frac{n_b}{t_s}\left[1 + \frac{t_s}{t_b}\right]} \tag{5}$$

$$\mathrm{MDA} = \frac{2.71}{\mathrm{F}t_s} + 2\mathrm{MSA} \tag{6}$$

When n_b is the background count rate, t_s and t_b are the count times for the sample and for the background, respectively.

2.8. Uncertainty and Statistic

Generally, the measurement uncertainty is the most straightforward means for dose estimation. In the situation where the activity level is low and closed to the detection limit, the counting uncertainty can dominate the overall uncertainty. The combine uncertainty was calculated as equation 7, calculation expanded uncertainty at 95% confidence (α=0.05) from URACHEM [5], where U_{rep} is the standard uncertainty from repeatability, U_{eff} is the standard uncertainty from efficiency calibration and U_{cps} is the standard uncertainty from the count per second.

$$u_c = x_i \sqrt{\left(\frac{u_{rep}}{x_i}\right)^2 + \left(\frac{u_{Eff}}{Eff}\right)^2 + \left(\frac{u_{cps}}{CPS}\right)^2} \tag{7}$$

3. Results

3.1. Calibration

The efficiency calibration fitted curves of two types of radiation detectors, NaI and HpGe detectors with Genie 2000 gamma spectroscopy software shown in figure 2.

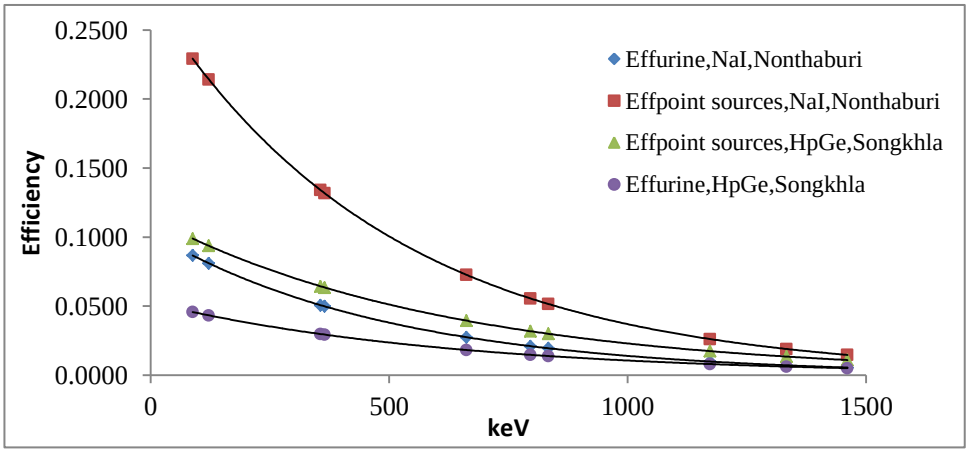

Figure 2. The efficiency calibration fitted curves for point sources and urine sample counting by NaI and HpGe detectors by assuming 100% gamma ray abundance.

3.2. Gamma analysis

The spectrum of each radionuclide was calculated as equation 2. Table 1, figure 2 and 3 show the average activity of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in eight locations.

Table 1. The baseline quantities of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in 30 ml of urinary excretions From Thai people.

location	Average activity (Bq)				detector
	^{131}I	^{137}Cs	^{134}Cs	^{40}K	
Chiangmai	0.04±0.03	0.25±0.13	0.25±0.14	7.41±6.83	NaI
Nonthaburi	0.48±0.17	0.40±0.14	0.52±0.19	9.99±6.55	NaI
Maha Sarakham	0.30±0.11	1.22±0.42	0.68±0.38	9.03±5.34	NaI
Loei	0.05±0.02	0.06±0.02	0.04±0.02	0.33±0.21	NaI
Khon Kaen	0.17±0.07	0.36±0.14	0.22±0.09	8.86±7.21	Hp(Ge)
Bangkok	0.07±0.03	0.28±0.10	0.06±0.11	3.05±2.09	Hp(Ge)
Songkhla	0.07±0.03	0.06±0.02	0.05±0.02	2.82±1.48	Hp(Ge)
Pattani	1.83±0.75	2.46±0.99	1.31±0.53	21.19±10.49	Hp(Ge)
average	0.19±0.17	0.43±0.41	0.30±0.25	6.45±3.08	

The continuous uncertainty can be calculated in equation 8.

$$S_y = \sqrt{S_a{}^2 + S_b{}^2 + S_c{}^2 + ...} \tag{8}$$

Where S_a, S_b, S_c, … are uncertainties from determination of average activities in each location. When using the one-way analysis of variance (ANOVA), the most of the average activities in eight locations are significant differences. Because each location is difference of diet and environment.

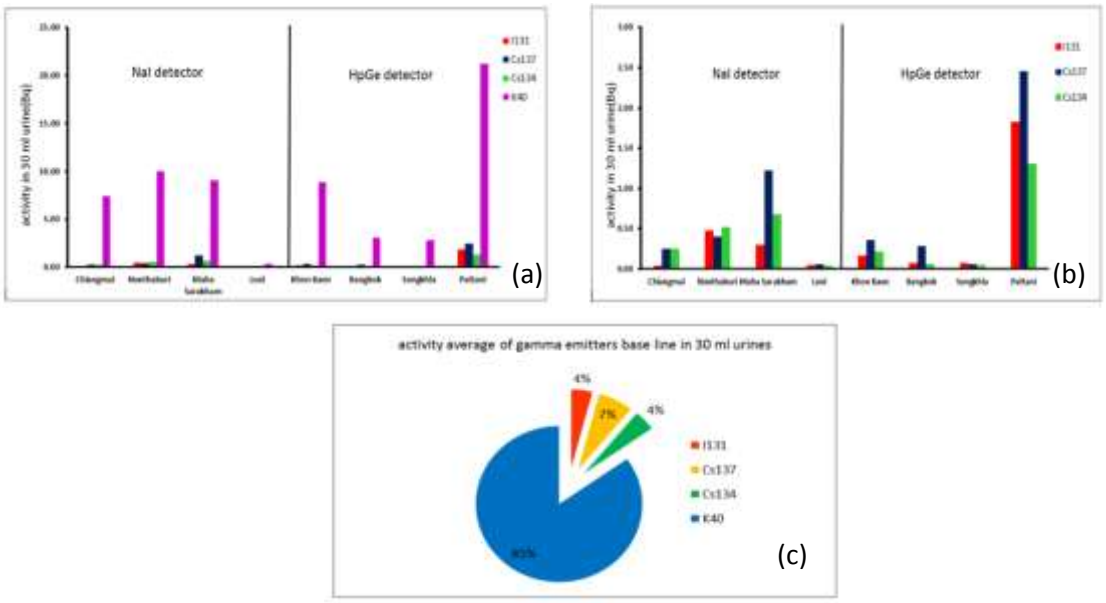

Figure 3. Radioactivity of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in 30 ml urine samples obtained from different locations of Thailand (a and b), the average activity is showed in (c).

3.3. Internal dose assessment

Due to the activities of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in urine samples are less than MDA, it means that the activities have no significantly in excess of the background level. The activities in 30 ml urine samples are not depend on the volume of urine. Therefore, the activity in daily excretion is equivalent to the activity from measurement. For ^{40}K, the predicted value of the measured quantity in urine from daily intake by ingestion equivalent to daily excretion because of its rapidly excretion after intake. The percentage of dietary potassium excreted in the urine was high. It is approximately 63 to 92% of intake from ref[4].

The estimation of intake can be calculated by assuming default parameter, absorption type F. Time after acute intake 1 day, intake by inhalation, the m(t) of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K, are 2.80E-01, 7.90E-03, 7.90E-03 and 6.90E-03 Bq per Bq intake, respectively. The intake by ingestion, the m(t) of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are 2.80E-01, 7.90E-03, 7.90E-03 and 6.90E-03 Bq per Bq intake, respectively. In addition, this research assumed that members of the public intake 50% by ingestion and 50% by inhalation. The dose coefficients for AMAD 1 μm for inhalation of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are 7.40E-09, 4.60E-09, 6.60E-09 and 2.10E-09 SvBq^{-1}, respectively. The dose coefficients for AMAD 1 μm for ingestion are 2.20E-08, 1.30E-08, 1.90E-08 and 6.20E-09 SvBq^{-1}, respectively. The committed effective dose equivalent due to intake of these radionuclides is shown in table 2.

Table 2. The committed effective dose equivalent in members of public due to intake of ^{131}I, ^{137}Cs,^{134}Cs and ^{40}K by assuming 50% acute intake by ingestion and 50% acute intake by Inhalation.

CEDE (mSv.y^{-1})			
^{131}I	^{137}Cs	^{134}Cs	^{40}K
2.36E-03±1.66E-03	1.15E-01±8.61E-02	1.16E-01±7.77E-02	9.44E-01±3.56E-01

3.4. Minimum detectable activity

The activities of ^{131}I, ^{137}Cs,^{134}Cs except for ^{40}K in 30 ml of urine, obtained from 8 locations of Thailand were lower than the MDA. It means that the activities have no significant in the excess of the background level, except in Pattani, which seems higher than the other. They showed that the activities are significantly in the excess of the background. The MDAs of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are shown in table 3.

Table 3. The MDAs of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K of urine sample counting in each location.

location	MDA (Bq)				detector
	^{131}I	^{137}Cs	^{134}Cs	^{40}K	
Chiangmai	0.88	1.39	1.60	50.81	NaI
Nonthaburi	0.88	1.39	1.60	50.81	NaI
Maha Sarakham	0.88	1.39	1.60	50.81	NaI
Loei	0.88	1.39	1.60	50.81	NaI
Khon Kaen	0.86	1.77	0.85	29.04	HpGe
Bangkok	0.40	0.50	0.73	9.76	HpGe
Songkhla	0.31	0.37	0.35	8.13	HpGe
Pattani	0.05	0.80	1.39	27.10	HpGe

4. Conclusion

This research shows that the average activities in 30 ml urines of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are 0.37 ± 0.09, 0.63 ± 0.13, 0.39 ± 0.08 and 7.84 ± 1.63 Bq, respectively. The average activities in eight locations are significant differences due to each location is difference of diet and environment when using the one-way analysis of variance (ANOVA). The intake of radionuclides and the assessed internal exposure dose in members of the public were calculated by assuming 50% intake by ingestion and 50% intake by inhalation. The committed effective dose equivalent of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K are 2.36E-03±1.66E-03, 1.15E-01±8.61E-02, 1.16E-01±7.77E-02 and 9.44E-01±3.56E-01 mSv per year, respectively. The internal dose, CEDE values are less than 1 mSv a year. These data from this research can be used as the baseline quantities of ^{131}I, ^{137}Cs, ^{134}Cs and ^{40}K in urinary excretions from Thai people and internal exposure dose. For further study, the radiation baseline quantities from this study can be applied as a reference data in case of nuclear accidents and can be used to study the long term health effect of internal contamination for Thai people.

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