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Comparative Study on U, Th and K Concentrations in Sediments at the Mae Moh Mine and Lam Phra Phloeng Dam

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ABSTRACT

The concentration of uranium, thorium and potassium in sediment matrices, collected from the Mae Moh mine in Lampang Province, Northern Thailand and from the Lam Phra Phloeng Dam on Nakhon Ratchasima Province, Northeastern Thailand were determined using gamma spectrometry together with a neutron activation analysis technique (NAA). A high purity germanium (HPGe) well-type detector and digital spectrometer were calibrated by reference standard sources to obtain high efficiency for the quantitative evaluation. The concentration of uranium, thorium, and potassium of the Mae Moh mine samples achieved by these techniques was in the range 0.08 to 1.40 ppm, 4.60 to 6.70 ppm, and 0.80 to 1.50%, respectively and significantly lower than the samples collected from the Lam Phra Phloeng Dam which were in the range 2.28 to 2.96 ppm, 8.43 to 16.01 ppm, and 2.04 to 3.30%, respectively. It was shown that the concentration of naturally occurring radioactive materials (NORM) depended on the type and formation age of the sediment.

Key words: HPGe, NAA, gamma-spectrometry, uranium, thorium, potassium, Mae Moh mine, Lam Phra Phloeng Dam

INTRODUCTION

Naturally occurring radionuclides of the uranium (^{238}U), actinium (^{235}U) and thorium (^{232}Th) series, and the radioactive isotopes of potassium (^{40}K) in raw building materials and products derived from rock and soil are sources of external and internal radiation exposure in dwellings. However, the contribution of actinium series isotopes to natural radiation is usually neglected due to very low concentrations of ^{235}U. The specific activities of ^{238}U, ^{232}Th and ^{40}K in raw building materials mainly depend on their geological sites of origin and their geochemical characteristics. Therefore, knowledge of radiation levels and basic radiological parameters in building materials is essential to assess possible radiological risks to human health. Normally, the external exposure is caused by direct gamma radiation originating from the above mentioned members of the radioactive series. Internal exposure of the respiratory tract to alpha particles

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is due to the radioactive inert gases, radon (^{222}Rn, a daughter product of ^{226}Ra) and thoron (^{220}Rn, a daughter product of ^{224}Ra), and their short-lived decay products, which are emitted from building materials into room air, (Hubert, 2001; Turhan and Gunduz, 2008) .

Several methods have been employed to determine the contents of ^{238}U, ^{232}Th and ^{40}K in archaeological and geological materials. Each method has its own merits and convenience. One of them is gamma-ray spectrometry and neutron activation analysis (NAA) (Ikeya, 1993).

The Mae Moh Mine in Lampang province, is well known for its lignite. It is located in the northern part of Thailand, in a stratigraphic bedding plane in steady sediment that dates geologically to about 3-31 million years ago (Ratanasthien, 2002). The Lam Phra Phloeng Dam is located in Nakon Ratchasima province in northeastern Thailand. Construction started in 1963 and was finished four years later, (Lorsirirat, 2007).

In this comparative study, samples from two areas (Mae Moh mine and Lam Phra Phloeng Dam) were analyzed using gamma spectrometry with an HPGe well-type detector and Gamma Vision-32 V 3.2 Gamma-Ray Spectroscopy software at the Office of Atoms for Peace (OAP) to determine the specific activity of ^{238}U, ^{232}Th and ^{40}K. One of the aims was to use the results as reference data that could assist in the assessment of potential radiological hazards related to these materials by computing the radium-equivalent activity and other factors, such as an activity index, the indoor gamma absorbed dose rate and the effective dose rate.

The Mae Moh Mine is situated in the Mae Moh District of Lampang province, which is about 26 km east of Lampang city. The mine is located in an area of about 135 km^2, 7 km in an east-west direction and 16 km north-south (Figure 1). The basin floor is about 320-340 m above mean sea level. The tertiary sediments have been named as the Mae Moh Group. The group consists of three formations, namely the Huai King, Na Khaem, and Huai Luang formations, in ascending order, with a total thickness of nearly 1,000 m (Figure 2) (Songthama *et al.*, 2005).

The Huai King formation consists of a sequence of upward grading conglomerate, sandstone and pebbly sandstone, siltstone and finally fining upwards into interbedded red and

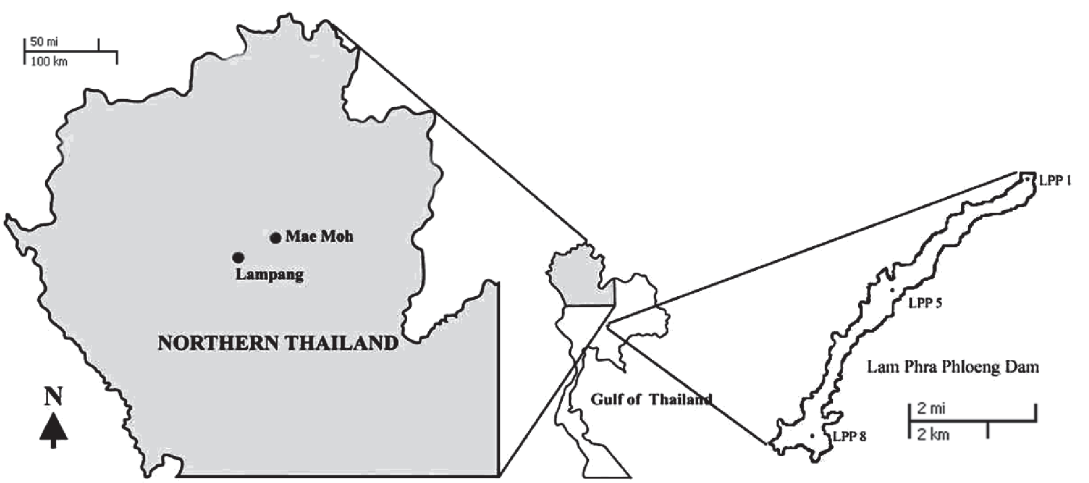

Figure 1 Map of Thailand showing location of the Mae Moh mine and Lam Phra Phloeng Dam.

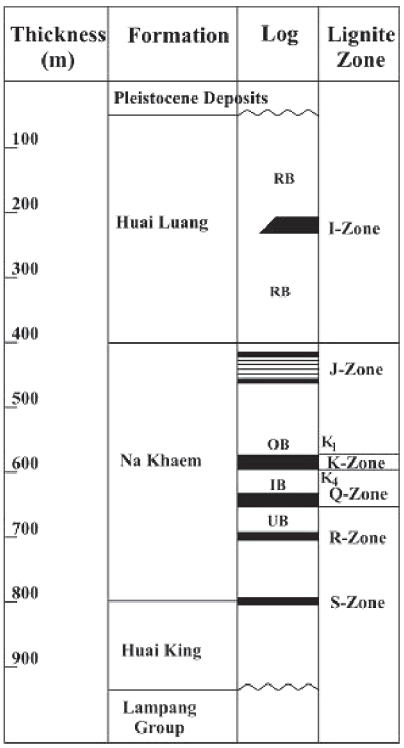

Figure 2 Example schematic of the lithostratigraphic units of the Mae Moh group.

gray claystone. The Na Khaem formation is a coal measure comprising three main coal zones, Q, K, and J assigned to the Middle Miocene age. This formation has been divided into three members as overburden (OB), interburden (IB) and underburden (UB). UB contains a greenish gray to gray claystone with a thin layer of coal, named the R coal zone. The IB is composed of two main coal zones, which are the Q coal zone in the lowermost part and the K coal zone in the uppermost part. A freshwater fossil shell bed nearly 12 m thick was discovered between coal zones K-3 and K-4 in 2003 by mine staff of the Mae Moh coalmine. The OB consisting of claystone with a series of coal layers is named the J coal zone. The Huai Luang formation is composed of claystone and siltstone with some sandstone and a red to brownish red color is the general characteristic of this formation. (Songthama *et al.,* 2005)

The Lam Phra Phloeng Dam is located in Nakhon Ratchasima province in Thailand (Figure 1) and is situated in Amphoe Pak Thong Chai on a side road about 28 km from the main highway. The dam length is about 11 km, the depth is 15-25 m and the total capacity is about 110 million m^3. The dam is 263 m above mean sea level (Lorsirirat, 2007).

The purpose of this study was to determine the concentrations of U, Th and K in archaeological samples from the Mae Moh mine confined within Redbed deposits (RB) from the Huai Luang formation and the gray claystone layer, quartz, fossil beds and surrounding sediment in coal between coal zones K-3 and K-4 from the Na Khaem formation. The same determination at the Lam Phra Phloeng Dam was confined within the crest (LPP 1), middle (LPP 5) and end of the dam (LPP 8) (Figure 1).

MATERIALS AND METHODS

Collection and preparation of samples

The sediment samples were collected from the Mae Moh mine and Lam Phra Phloeng Dam. In the Mae Moh mine, a sample was collected from the Huai Luang formation and six other samples from the Na Khaem formation. At the Lam Phra Phloeng Dam, three samples were collected from the crest (LPP 1), middle (LPP 5) and end of the dam (LPP 8). All samples were oven dried at 60°C for 24 h, crushed, pulverized to a fine powder and homogenized. Each powdered sample of 300 to 400 mg was accurately weighed and placed in polyethylene vials with the same geometry and volume as the vials containing 600-700 mg of a standard reference material (NBS SRM 2709: San Joaquin Soil). Then, the vials were sealed and left for about one month to allow the standards and the samples to reach secular equilibrium, following the method of Soliman (2006).

Neutron irradiation

All samples were irradiated in the Thai Research Reactor-1/Modification (TTR-1/M1) at the Office of Atoms for Peace (OAP) in two steps. Firstly, long time irradiation with an epithermal neutron flux of 2×10^9 n/(cm^2.s) for 12 h and a cooling time of 5-6 d were used to determine the concentration of U and Th. The second, short time irradiation with a thermal neutron flux of 2×10^{11} n/(cm^2.s) for 10 min and cooling for 12 h were used to determine the concentration of K.

Gamma-Ray measurement

The gamma spectrometer system consisted of a GWL series HPGe (high-purity germanium) coaxial well with 0.5 mm aluminum absorbing layers (well wall) thickness, mounted in a vacuum-tight cryostat (Model GWL-120230, crystal diameter 54.9 mm, well with inside diameter 10 mm and an active well depth of 40 mm), a liquid-nitrogen Dewar and dipstick cryostat (Model HJ-GWL) and 1500 volts high voltage supply. A computer-based MCA, (DSPEC) and GammaVision-32 V 3.2 Gamma-Ray Spectroscopy software, with a graphical user interface, that was ideal for the manipulation and analysis of spectra with a personal computer was used. Four isotopes (Ba-133, Eu-152, Cs-137 and Co-60) were used for energy and efficiency calibrations. After irradiation, the γ-radiation spectra of the samples and standard soils were measured and compared using the flux monitors, which then gave a quantitative ratio correction of the gamma-ray spectrum.

Due to the short half-life of ^{239}U and ^{233}Th, which are the activation products of ^{238}U and ^{232}Th, respectively, it was convenient to measure their beta decay isotopes, ^{239}Np and ^{233}Pa, as shown in Equations 1 and 2, respectively (Rossini *et al*., 1991).

$$^{238}U(n,\gamma)^{239}U \xrightarrow{\beta-} {}^{239}Np(2.35d) \quad (1)$$

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow{\beta-} {}^{233}Pa(27d) \quad (2)$$

RESULTS AND DISCUSSION

Gamma spectra of U, Th and daughters are shown in Figure 3a and those of K are shown in Figure 3b. The concentrations of U, Th and K, obtained from seven different samples (from the Mae Moh mine), were in the range 0.02 to 3.05 ppm, 0.04 to 11.00 ppm, and 0.01 to 1.53%, respectively. The concentrations of U, Th and K, obtained from seven different samples (at the Lam Phra Phloeng Dam), were in the range 1.50 to 3.00 ppm, 2.80 to 16.90 ppm, and 2.00 to 3.60 %, respectively (Table 2).

CONCLUSIONS

The present experiment showed that gamma spectrometry together with NAA techniques is well suited to determine the concentration of U, Th and K in the sediments from the Mae Moh mine and Lam Phra Phloeng Dam. Due to the formation of freshwater fossils and surrounding sediments, the naturally occurring

Table 1 The important nuclear data of isotopes derived from neutron activation of U, Th and K (Rossini *et al*., 1991; Ghawi *et al*., 2005; Soliman, 2006).

Element	Isotope	Half-life	Energy of emitted gamma ray (keV)	% of emission
U	^{239}U	23.5 min	74.6	59.3
	^{239}Np	2.55 d	277.6	14.1
Th	^{233}Th	22.3 min	86.5	2.6
	^{233}Pa	27.4 d	311.9	33.7
K	^{42}K	12.36 h	1524.7	17.9

radionuclides gave small values when compared to concentrations in other embedded strata. Since these fossil shells were collected from a thick bedding plate almost 12 m thick, surrounded by many small, fractional, freshwater fossil shells, the concentration can be considered acceptable. The naturally occurring radionuclides in the sediments from the Lam Phra Phloeng Dam had more concentrated values than from the Mae Moh mine because their sediment characteristics differed. In addition, the sediments were from different eras, with the sediments from the Mae Moh mine from the tertiary era, while the others from the dam were from the quaternary era. The results could be used as reference data and could be used to assess the potential radiological hazards by computing the radium-equivalent activity.

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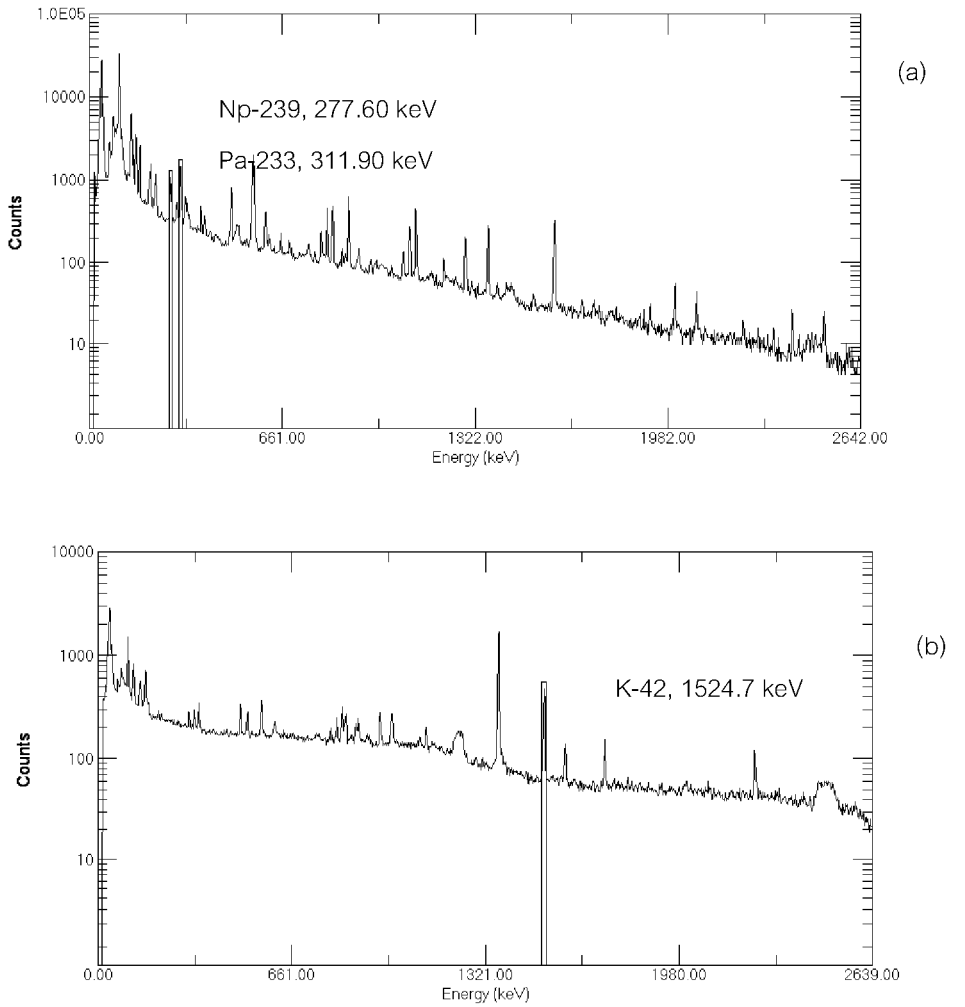
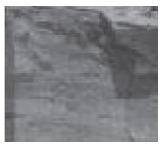


Figure 3 Measurement of the main part of a typical gamma ray spectrum of sample MMm 6sr (No.6 from the Huai Luang formation). The important identified photopeaks and their associated radionuclides are shown with gamma spectra for radioactive elements (a) U, Th and (b) K.

Table 2 Concentrations of U, Th and K of seven samples from Mae Moh mine and three samples from Lam Phra Phloeng Dam.

No.	sample	Characteristic of sample	^{238}U (ppm)	^{232}Th (ppm)	^{40}K (%)
1	MMm 1		0.80 ± 0.04*	6.20 ± 0.28	1.20 ± 0.05
2	MMm 2		0.97 ± 0.04	4.60 ± 0.21	0.80 ± 0.04
3	MMm 3		1.40 ± 0.06	6.70 ± 0.31	1.50 ± 0.07
4	LPP 1		2.28 ± 0.10	8.43 ± 0.39	2.04 ± 0.09
5	LPP 5		2.58 ± 0.12	14.35 ± 0.66	2.18 ± 0.10
6	LPP 8		2.96 ± 0.14	16.01 ± 0.73	3.30 ± 0.15

* total uncertainty at 95% confidence interval.

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