Radioisotope Determination in Sediments in Lam Phra Phloeng Dam, Northeastern Thailand, Using Gamm Ray Spectrometry

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ABSTRACT

The Lam Phra Phloeng dam in Nakhon Ratchasima province is under severe reduction of water caused by deforestation and agriculture. The Pb-210 activities of a core of the dam's sediment were measured using gamma spectrometry. These Pb-210 activities were used to determine the rate of sedimentation of the core. The sedimentation rate, determined using the constant rate of supply model, averaged 0.63 ± 0.10 gram per square centimeter per year. The Pb-210 dating method is the principal dating technique for very young sediments.

KEYWORD: Lam Phra Phloeng dam, gamma spectrometry, Pb-210, sedimentation rate, dating

INTRODUCTION

Lam Phra Phloeng dam is located in Nakhon Ratchasima province in northeastern Thailand (Figure 1). The dam was constructed in 1963 and began operating 4 years later. It is now one of the dams most seriously affected by sediment accumulation.

Deforestation in the dam's upper catchments reduced the forest area from 531 square kilometers in 1974 to 160.25 square kilometers in 1985, a reduction of 51.25 percent in 10 years (Lorsirirat, 2007). This was a rapid change from forest to agricultural. Tilling of land after harvesting makes it sensitive to sheet erosion. Periodically, the dam's region suffered from floods, as well as droughts. The increased erosion since forest clearance has led to an increase in sediment load in rivers draining to the dam's reservoir. As a direct result, the reservoir of the Lam Phra Phloeng dam is very shallow where major streams enter.

All these natural and human manipulations accelerated the sedimentation rate in the dam's reservoir. The sedimentation rate determines the effective life of the dam and knowing this is essential to the long term future of people who now benefit from the dam and its reservoir. Artificial and natural radioisotopes like Cs-137 and Pb-210 provide useful information as tracers to generate reasonable age estimates for sediments and, thus, allow determination of sedimentation rates (Walling, 2004). Determination of Pb-210 chronologies and sedimentation rate was originally developed by Goldberg in 1963, although its first application to lake sediments was by Krishnaswamy and others in 1971 (Gale and others, 1995).

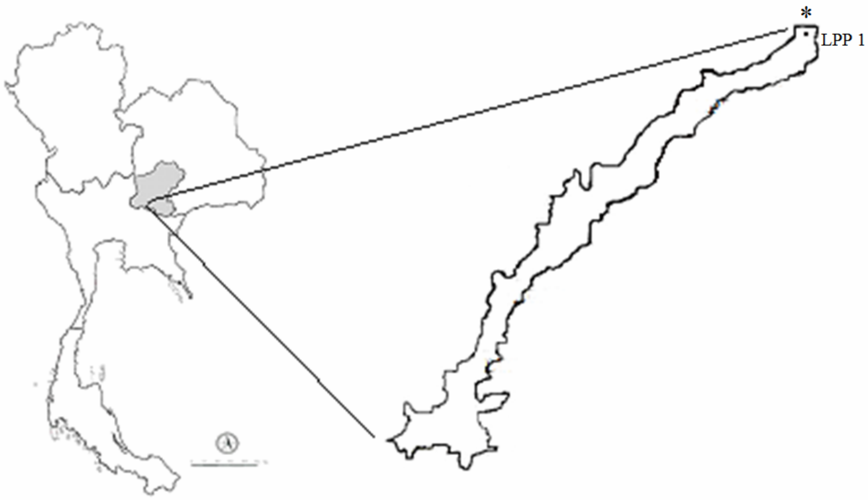

Figure 1. Map of Lam Phra Phloeng dam reservoir and sample location; asterisk is dam crest.

Cs-137 interacts strongly with micaceous clay minerals in soils and sediments (Comans and others, 1989). Cs-137 with a half-life of 30.2 years is produced by nuclear fission and has been released to the environment as a result of nuclear weapon testing during the 1950s to the 1970s. Its maximum atmospheric input was in 1963 and from the major accident at Chernobyl in 1986. However, nuclear weapon-derived Cs-137 inputs were significantly lower in the Southern Hemisphere than in the Northern Hemisphere (Zhang and Walling, 2005). Also, Cs-137 inputs in equatorial areas were considerably less than in the mid-latitude areas of Europe and North America (Walling and He, 1999). The low Cs-137 inventories associated with reduced receipt of fallout in these areas introduced measurement problems in terms of both detection limits and the long count times required to obtain results with an acceptable degree of precision.

Pb-210 with a half-life of 22.26 years continuously settles onto soil and sediment surfaces. It is a natural product of the U-238 decay series. It is derived from the decay of gaseous Rn-222, the daughter of Ra-226. Ra-226 occurs naturally in soils and rocks and generates Pb-210, which is in equilibrium with its parent (Zapata and Garcia-Agudo, 2000). On the other hand, Rn-222 gas produced from the decay of Ra-226 can diffuse and produce Pb-210 in the atmosphere. As fallout, some radionuclide Pb-210 is rapidly and strongly adsorbed on the soil surface. This soil can later be redistributed within sediments (Zapata, 2003).

In the study, environmental Pb-210 dating techniques were used to determine recent sedimentation rates. The total Pb-210 activity in sediment has two components. The first is a minor part that is in equilibrium with Ra-226 fixed to sediment. The second part is Pb-210 that is associated with the particulate matter. This Pb-210 is called unsupported Pb-210, it being in excess, or unsupported. This unsupported Pb-210 is formed in the atmosphere after Rn-222 decay and is deposited on the surface with particle materials. The exponential decrease of the accumulated unsupported Pb-210 can be used to estimate sediment accumulation rate (Robbins, 1978). The required measurements of Pb-210 and Ra-226 activity can be made conveniently by direct gamma spectrometry using a low-energy and low background high-purity germanium detector.

MATERIALS AND MEASUREMENT

Sediment Samples

The PPL 1 sediment cores were taken in September, 2006, using a gravity corer with a 10-kilogram weight. The corer's PVC tubes were 5.4 centimeters in diameter and 75 centimeters long. After recovery, the sediment cores were sliced into segments 1 centimeter thick and these segments were stored in zip plastic bags. The sediment rim of each slice was removed and discarded to avoid contamination. The sediment slices were frozen at 4^{o}C and transported in an ice box. In the laboratory, each slice was divided into sub-samples to determined particle size, organic matter content, and radioactivity. For particle size determination, the samples were stored in a refrigerator at 4^{o}C until used. The rest of the samples were weighed and dried at 60^{o}C until the sediment weight was constant. The samples were pulverized in a centrifugal ball mill and sieved through a 125 micrometer stainless steel sieve to achieve a uniform particle size.

Gamma spectrometric analysis for Pb-210 and Ra-226 in sediment samples

Pb-210 was determined using its gamma emissions at 46.5 kiloelectron volts. Ra-226 was determined by the 351.9 and 609.3 kiloelectron volt gamma rays emitted by its daughter isotope Pb-214 and Bi-214 after 3 weeks storage in sealed containers to allow radioactive equilibrium between Ra-226 and its daughter Rn-222. In this method, about 2 grams of sediment was placed in an ampoule having a 10-millimeter inside diameter and a height of 30 millimeters, the ampoule then being weighed and sealed. Each sample was measured more than 50,000 seconds to obtain good statistical results. These results were then analyzed using the GammaVision-32 V 3.2 gamma ray spectroscopy software. The gamma ray method requires the preparation of standard and background samples. The radioisotopes Ba-133, Eu-152, Cs-137 and Co-60 were used for energy and efficiency calibration. In addition, the precision and accuracy of the method were determined by analyzing certified reference materials, IAEA-327. The activities of the natural radionuclide in the standard sample are listed in Table 2.

Table 1. Core LPP 1, sediment properties, age, and sedimentation rate.

Depth (cm)	Unsupported Pb-210 (Bq/kg)	Clay %	Mud %	Cumulative Dry Mass (g/cm2)	Age (y)	Sedimentation Rate ($g/cm^2/y$)
0 - 1	67.92 ± 12.62	4.35	68.56	0.15 ± 0.15	0.25 ± 0.37	0.61 ± 0.91
1 - 2	55.38 ± 9.65	7.37	73.41	0.46 ± 0.15	0.73 ± 0.51	0.63 ± 0.43
2 - 3	59.92 ± 11.55	6.00	70.39	0.80 ± 0.16	1.24 ± 0.61	0.65 ± 0.32
4 - 5	52.66 ± 9.72	5.02	66.24	1.55 ± 0.17	2.35 ± 0.78	0.66 ± 0.22
6 - 7	46.73 ± 8.14	4.43	68.18	2.39 ± 0.18	3.50 ± 0.87	0.68 ± 0.17
7 - 8	51.67 ± 9.55	6.02	70.89	2.87 ± 0.19	4.16 ± 0.90	0.69 ± 0.15
8 - 9	43.52 ± 7.86	5.41	72.40	3.37 ± 0.20	4.84 ± 0.93	0.70 ± 0.13
10 - 11	54.31 ± 9.83	5.28	71.52	4.38 ± 0.21	6.31 ± 1.03	0.69 ± 0.11
11 - 12	50.93 ± 8.41	5.22	72.16	4.89 ± 0.21	7.15 ± 1.05	0.68 ± 0.10
12 - 13	50.00 ± 7.69	4.75	68.66	5.42 ± 0.22	7.98 ± 1.06	0.68 ± 0.09
13 - 14	53.97 ± 8.57	4.59	69.84	5.94 ± 0.22	8.86 ± 1.08	0.67 ± 0.08
14 - 15	51.49 ± 7.73	4.72	69.38	6.46 ± 0.22	9.78 ± 1.10	0.66 ± 0.07
15 - 16	53.63 ± 7.71	3.42	71.94	6.98 ± 0.23	10.72 ± 1.12	0.65 ± 0.07
16 - 17	44.21 ± 6.81	4.37	69.00	7.49 ± 0.23	11.60 ± 1.14	0.65 ± 0.06
17 - 18	50.70 ± 8.10	4.29	65.84	8.02 ± 0.23	12.51 ± 1.16	0.64 ± 0.06
18 - 19	47.45 ± 6.97	4.63	67.34	8.55 ± 0.23	13.49 ± 1.18	0.63 ± 0.06
19 - 20	51.09 ± 8.18	3.31	73.18	9.04 ± 0.23	14.43 ± 1.20	0.63 ± 0.05
21 - 22	40.39 ± 7.58	4.23	73.91	10.03 ± 0.23	16.26 ± 1.24	0.62 ± 0.05
23 - 24	42.63 ± 7.50	4.10	72.64	11.07 ± 0.24	18.09 ± 1.29	0.61 ± 0.04
27 - 28	47.70 ± 8.56	4.46	69.85	13.16 ± 0.24	22.53 ± 1.38	0.58 ± 0.04
29 - 30	46.51 ± 7.47	4.68	68.65	14.24 ± 0.24	25.21 ± 1.43	0.56 ± 0.03
31 - 32	40.32 ± 7.77	3.50	67.37	15.32 ± 0.24	27.88 ± 1.47	0.55 ± 0.03
33 - 34	49.31 ± 7.92	4.76	70.71	16.38 ± 0.24	30.83 ± 1.52	0.53 ± 0.03
35 - 36	47.62 ± 8.21	5.48	78.22	17.41 ± 0.25	34.27 ± 1.58	0.508 ± 0.023

Table 2. Activity of natural radioisotopes in the standard sample for gamma spectrometry of sediment.

	Activity of Radionuclide ($Bq\ kg^{-1}$)		
IAEA-327	Pb-210	Ra-226	K-40
Value from IAEA Certificate	39.17	33.91	621.00
Measured value at OAP	39.94 ± 6.30	33.80 ± 5.33	619.23 ± 97.71

The gamma ray spectrometry system was a GWL series high-purity germanium coaxial well with 0.5-millimeter thick aluminum absorbing layers mounted in a vacuum-tight cryostat. The cryostat was model GWL-120230, with a crystal diameter of 54.9 millimeters, a well inside diameter of 10 millimeters, and an active well depth of 40 millimeters with a liquid-nitrogen Dewar dipstick cryostat, model HJ-GWL, and 1,500 volts high voltage supply which had an efficiency of 27 percent. The computer-based multichannel analyzer used had 16,383 channels and a gain function of 0.75 kiloelectron volts per channel. The resolution of the 1.33 megaelectron volt gamma ray peak from Co-60 is 2.30 kiloelectron volts and the integrated background from 25 kiloelectron volts to 2 megaelectron volts is 1.4 counts per second. This method has the advantage of being non-destructive and the sediment samples can be used for other measurements. The gamma ray spectrometry was done at the Office of Atoms for Peace.

MODEL CALCULATIONS

The Pb-210 sedimentation rate was determined using the constant rate of supply model that was originally proposed by Goldberg in 1963 and developed by Appleby and Oldfield in 1978 (Appleby and Oldfield, 1978; Gale and others, 1995). This model is based on the assumption that there is a constant flux of unsupported Pb-210 into a reservoir over its entire life. The age of sediment of depth, *x*, is given by the ratio of the unsupported Pb-210 activity below that point to the total unsupported Pb-210 in the sediment column:

$$t = \frac{1}{\lambda} \ln \frac{A_c(0)}{A_c(x)} \quad (1)$$

where, $A_c(0)$ is the total activity of unsupported Pb-210 in the sediment column, $A_c(x)$ is the accumulated activity of Pb-210 below depth x, t(y) is the age of the layer at depth x, λ is decay constant, 0.03114 y^{-1}.

RESULTS AND DISCUSSION

Core LPP 1 is 36 centimeters long and its sedimentation rate estimated from Pb-210 varied from 0.50 to 0.70 gram per square centimeter per year. Its average sedimentation rate was 0.63 ± 0.10 gram per square centimeter per year. Unsupported Pb-210 activity ranged from 40.32 to 67.92 becquerel per kilogram. Table 2 summarizes the core's sediment properties. Figure 2 shows the age of sediment versus depth for the core LPP 1.

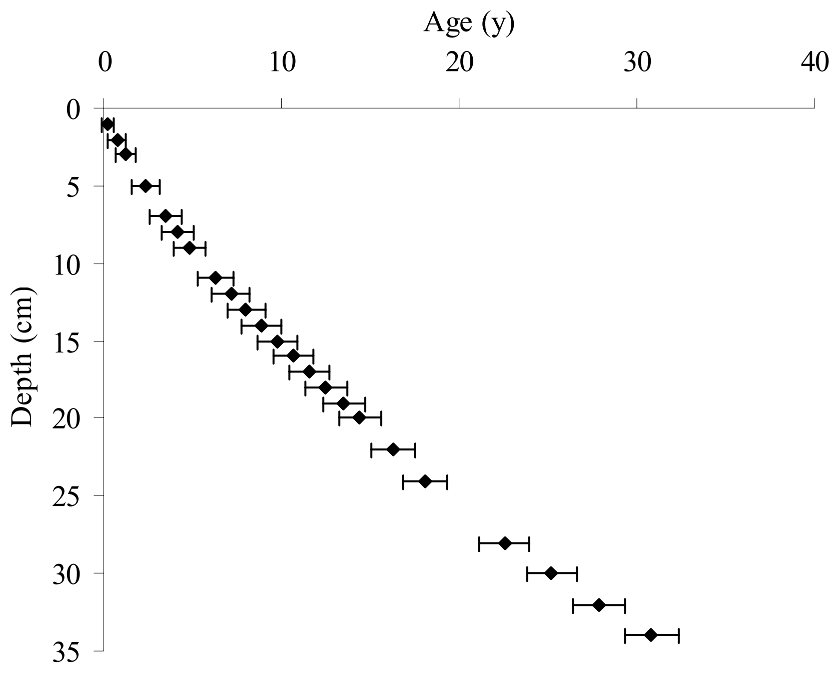

Figure 2. Core LPP 1, age of sediment versus core depth; length of bar indicates possible age error.

The unsupported Pb-210 activity was variable because the Pb-210 that was deposited on land can be dissolved and discharged to water resources nearby and, thus, added to Pb-210 that is in water due to the decay of Ra-226. The natural unsupported Pb-210 in the atmosphere can be deposited on the surface everywhere. Some portion of this may accumulate on land while the other portions can fall onto water. Pb-210 on land can be transferred into water and after being suspended for a moment can mix with Pb-210 that falls from the atmosphere. This increases the activities of the Pb-210 mass in the water, the amount of increase depending on the water in-flow velocity, particle size, gravitational force, and such factors as bottom slope.

CONCLUSION

The sedimentation rate and dating at Lam Phra Phloeng dam were studied using radionuclide. This was the first time gamma spectroscopy was used for direct measurement of Pb-210. Gamma spectrometry may be the only method needed for Pb-210 measurement. It also reduces the need for laborious chemical treatment of samples. Sedimentation rate and dating were done using the constant rate of supply model. The average sedimentation rate in the Lam Phra Phloeng dam reservoir is estimated to be 0.63 ± 0.10 gram per square centimeter per year. The study data show a drastic change in sedimentation conditions that are likely connected to rebuilding of the dam. The observed unsupported Pb-210 activities were low and did not decrease exponentially with depth. They could have been caused by several factors.

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