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Environmental Assessment of Natural Radioactivity in Soil Samples from the LUSI Mud Volcano, Indonesia

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Abstract

The environmental impacts and hazards due to the unstoppable hot mud flow by the East Java 'LUSI' Mud Volcano are increasing since its unexpected eruption on May 29, 2006. Analysis should be undertaken, not only to examine its impact on human health and the environment, but also to explore the potential benefits of the mud flow. One may be able to tap the mud flow as a material source for brick and cement. Recently there has been great concern about the health risks associated with exposure to natural radioactivity present in soil and building materials all over the world. In this context, measurements for natural radioactive isotopes such as ^{238}U and ^{232}Th series, and ^{40}K in mud samples were carried out using the HPGe (High-Purity Germanium) detector to determine the re-usability of the mud. ^{226}Ra, ^{232}Th and ^{40}K activity concentrations were found to be 13±1, 15±1 and 111±3 Bq/kg (1 Bq = 1 sec^{-1}), respectively, and the corresponding activity index was found to be 0.16±0.02. These values were compared with previous data and our measured accuracy was improved by a factor of nine at the maximum. Radium equivalent activity, external and internal hazard indices, and annual effective dose equivalent were also evaluated and all were found to be within acceptable limits.

Keywords: gamma ray spectrometry; LUSI; mud volcano; radioactivity; building material

1. Introduction

A two-year-old mud volcano with an eruption site named 'LUSI' (Lumpur "mud"-Sidoarjo), started to form in East Java, 30 km south of Surabaya, on May 29, 2006. Already more than seven square kilometers in area, it is still flowing at 100,000 cubic meters per day. That is enough to fill 53 Olympic swimming pools or submerge a football field under 610 feet of mud; the depth of a 61-story building. It has displaced more than 30,000 people and has already cost Indonesia $3.7 billion in damages and damage control (Cyranoski, 2007) (Figs. 1 and 2). The trigger mechanisms are still a subject of debate. They include: a) the Yogyakarta earthquake of May 27, 2006 - a magnitude of 6.3, approximately 250 km to the southeast, causing almost 6,000 deaths; b) the drilling of a 2,800-meter-deep gas exploration well (150 m away) by Lapindo Brantas; and c) a combination of the earthquake and drilling operations (Mazzini *et al*., 2007; Davies *et al*., 2008).

A disaster of this enormity dictates that scientific studies be undertaken to examine its natural and man-made triggering mechanisms, and to devise emergency response strategies to stem the flow, to manage social impacts, and to minimize environmental impacts and hazards. The Indonesian government may be able to tap the mud flow as a source of material for brick and cement, use it as a source of geothermal energy to produce electricity, or use it for therapeutic skin treatment if the sludge is determined to be non hazardous. The government has made many attempts to stem the flow of mud from the source, including dropping beach ball-sized concrete balls into its mouth, and building dams to channel the sludge to sea. Unfortunately the flow rate does not seem to be affected by these trials and recent heavy rains in early January caused a breach in the levees, forcing more than a hundred families to evacuate.

In addition, almost 12,000 medical treatments have been carried out, mainly for people affected by the release of hydrogen sulphide gas, which has reached concentrations of 700 ppm. The Indonesian Ministry of Environment requested technical assistance from the United Nations Office for Coordination of Humanitarian Affairs (OCHA). The United Nations Disaster Assessment and Coordination (UNDAC) team was deployed to determine environmental impacts/management of the mud. During the mission from June 25 to July 6, and also on July 27, 2006, ^{226}Ra, ^{232}Th, and ^{40}K concentrations in mud samples were measured to determine the re-usability of the mud in terms of building materials (UNDAC, 2006). Radioactive isotopes are associated with geological formations and could occur as Naturally Occurring Radioactive Materials (NORM). The main source of the erupted mud can be constrained between ~1615-1828 m in depth (Mazzini *et al*., 2007), which is Pleistocene bluish gray clay in the era of 1.8 million to 10,000 years BP.

Figure 1. The LUSI (Lumpur mud-Sidoarjo) mud volcano eruption site in the Porong area, East Java, Indonesia (Sidoarjo archive, 2006).

The purposes of this study were to assess the re-usability of the mud for brick and cement by examining it not only for ^{226}Ra, ^{232}Th, and ^{40}K, but also for the entire radionuclide identified in the γ-ray energy spectrum using a large volume and low-background HPGe spectrometer, and periodic monitoring for aspects of NORM on samples taken about 8 months apart.

2. Materials and Methods

2.1. Sampling

Our sampling was carried out at the LUSI site on March 12, 2007 in cooperation with the Sepuluh November Institute of Technology of Surabaya (ITS). The sample was then packed in a standard plastic marinelli container, hermetically sealed and stored for about four weeks prior to counting, so as to ensure radioactive equilibrium between ^{226}Ra and its short lived progeny. The sample was measured directly, without any chemical or physical treatments, at the position closest to the aluminum end-cap of the detector that has a thickness of 1.0 mm.

Figure 2. Upper: Location of the LUSI mud volcano in East Java. Lower: Satellite photo from August 28, 2008. Satellite photo courtesy of CRISP (CRISP 2008). Image size is approx. 3.795 km x 4.036 km.

Table 1. Specifications of the HPGe detector

Manufacturer	EG&G ORTEC
Detector model	GEM-110225
Crystal size	
Diameter × Length (mm)	80.6 × 103.1
Volume (cm^3)	526
Relative efficiency at 1.33 MeV	112%
Resolution	
at 1.33 MeV ^{60}Co	1.92 keV
at 122 keV ^{57}Co	0.83 keV
Peak-to-Compton ratio	
for 1.33 MeV ^{60}Co	90.3
Lineshape	
FWTM/FWHM	1.92
FWFM/FWHM	2.67

2.2. γ-ray measurement by low-background and large volume HPGe spectrometer

The low-background HPGe spectrometer was used for the γ-ray measurement. The spectrometer was a large volume, 526 cm^3 coaxial type HPGe detector with 10 cm lead, and 5 mm copper shields, located at the Radiation Research Facility in Higashi-Hiroshima. The specifications of the HPGe detector are summarized in Table 1.

The measured energy spectra for the sample and no sample (background) for the live time of 80,000 sec each are shown in Fig. 3. The detector was calibrated using the natural radioactive back-

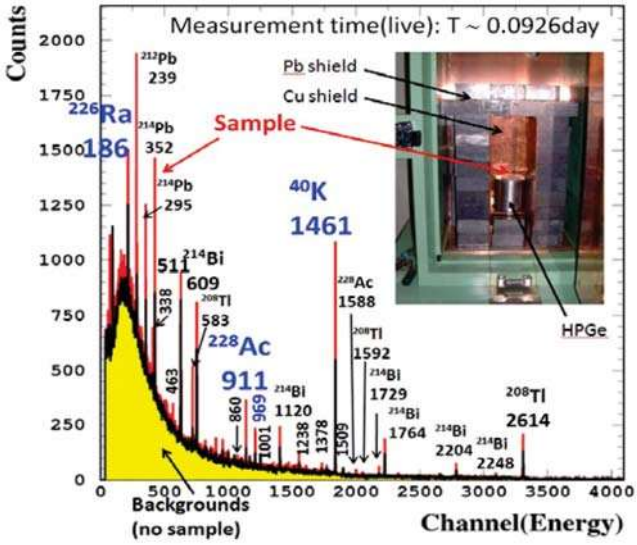

Figure 3. γ-ray spectra of LUSI soil sample and background measured with 526 cm^3 HPGe detector.

Table 2. Results from γ measurements in Bg/kg

Sample ID	A	B	C	D	E	Our result
Weight (g)	142	146	338	181	150	43
^{226}Ra (Bq/kg)	49±8	61±10	21±2	11±5	19±7	13±1
^{232}Th (Bq/kg)	48±8	86±10	35±3	13±4	22±5	15±1
^{40}K (Bq/kg)	740±140	580±130	200±30	180±50	120±40	111±3
Activity Index (*I*)	0.65±0.07	0.83±0.08	0.32±0.02	0.16±0.03	0.21±0.04	0.16±0.02

ground itself. The ^{226}Ra activities could be ascertained using the photo peaks of its daughters: ^{214}Pb (295 keV, 352 keV) and ^{214}Bi (609 keV, 1120 keV, 1764.5 keV) that were in secular equilibrium with ^{226}Ra following four weeks storage, due to the difficulty of direct measurement caused by the overlap with ^{235}U at 186 keV. In the case of ^{232}Th, the photo peaks of ^{228}Ac (338 keV, 911 keV, 969 keV), as well as ^{208}Tl (583 keV, 860 keV, 2614.6 keV) were measured. ^{40}K was measured directly from its 1461 keV γ-ray peak. Each activity contamination *A* (Bq/kg) was obtained by the following formula:

$$A = \frac{N}{T \times \varepsilon_{\gamma} \times I_{\gamma} \times W} (\text{Bq/kg}) \qquad (1)$$

where *N* is net peak counts (background subtracted), *T* is the measurement time (sec), ε_{γ} is the detection efficiency of the HPGe detector evaluated by GEANT4 (Agostinelli *et al*., 2003), I_{γ} is the branching ratio of the γ emission for the decay mode, and *W* is the sample weight (gram). As anticipated, the anthropogenic (artificial) radionuclide ^{137}Cs (from nuclear weapons testing and Chernobyl fallout) was undetectable. However, another anthropogenic radionuclide ^{125}Sb (427.9 keV, 463.4 keV) was detected and its activity concentration is about 1 Bq/kg. Further study will be needed to verify and clarify its origin.

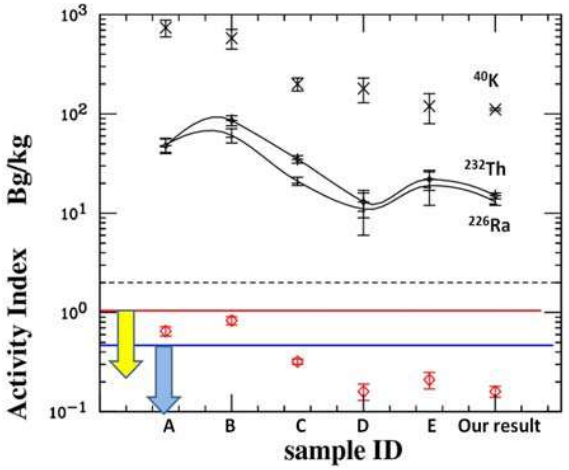

Figure 4. Activity concentrations (Bq/kg) and activity indices (I) of the LUSI soil samples. The lower limits of regulatory control are shown in the vertical lines for 1 mSv and 0.3 mSv, respectively.

3. Results and Discussion

3.1. Activity concentration index and comparison with European Commission Guidance

Because more than one radionuclide contributes to the dose, it is practical to present investigation levels in the form of an activity index. The European Commission Guidance document proposes the introduction of an activity concentration index (*I*), used to access the safety requirement of building materials:

$$I = \frac{A_{Th}}{300(\text{Bq/kg})} + \frac{A_{Ra}}{200(\text{Bq/kg})} + \frac{A_{K}}{3000(\text{Bq/kg})} \qquad (2)$$

where A_{Th}, A_{Ra}, and A_{K} are the thorium, radium, and potassium activity concentrations (Bq/kg), which were obtained by Eq. 1. Our result is compared with the previous results (UNDAC, 2006) and summarized in Table 2 and Fig. 4. It is noted that the radionuclide concentration of natural origin in LUSI mud is lower than the world average for building materials (^{226}Ra = 50 Bg/kg, ^{232}Th = 50 Bg/kg, ^{40}K = 500 Bq/kg) except for sample A and B. Similarly, population weighted averages in soils are: ^{238}U = 33 Bq/kg, ^{232}Th = 45 Bq/kg, ^{40}K = 420 Bq/kg (Somlai *et al*., 2008).

The activity index should not exceed the values in Table 3, depending on the dose criterion and the manner in which, and how much of the material is used in a building (ECRP 112, 1999). Regulatory control should be considered for materials that give rise to doses between 0.3 mSv and 1 mSv per year

Table 3. The relationship between the activity index (*I*) and received dose per year

Dose criterion	< 0.3 mSv	< 1 mSv
Bulk Materials, eg. concrete	$I \leq 0.5$	$I \leq 1$
Superficial and other materials with restricted use: tiles, boards, etc.	$I \leq 2$	$I \leq 6$

(Sv = W_R xGy (1 Gy = 1 J/kg), where W_R is the radiation weighting factor: ex. W_R = 1 for γ, X, and β-ray, W_R = 20 for α-ray, etc). Materials giving doses below 0.3 mSv should be exempt from all restrictions and those above 1 mSv must be regulated.

All of the samples analyzed had $I \leq 1$. This means that the mud could be used as bulk or superficial material without regulation, as occupants of buildings manufactured from such substances would receive an annual dose of less than 1 mSv. It is noted that two samples exceeded the lower limit of $I \leq 0.5$ which could give rise to doses of 0.3 mSv/year. Regular monitoring would be advised, in addition to monitoring for heavy metals and organic compounds. It is also emphasized that our measured accuracy is improved by a factor of 1.3 to 9 in virtue of the large volume detector and thorough background subtracted analysis, in spite of its small sample weight (about 1/5).

3.2. Radium equivalent activity

It would be useful to check exposure due to the γ radiation, which is defined in terms of the radium equivalent activity Ra_{eq} using the following equation (Faheem *et al.*, 2008):

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \leq 370 \qquad (3)$$

According to this formula, 1 Bg/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th, and 13 Bq/kg of ^{40}K yield the same γ-ray dose. The obtained radium equivalent activity is 43±4 Bq/kg, which is well below the upper limit of 370 Bq/kg.

3.2. Dose in air, annual effective dose, and the corresponding external and internal hazard indices

In order to evaluate the dose rate in air, the following formula was used:

$$\dot{D} = \sum_x A_x \times C_x \qquad (4)$$

where A_x (Bq/kg) is the activity of ^{226}Ra, ^{232}Th, and ^{40}K, as calculated by Eq. 1, and C_x (nGy/hour per Bq/kg) is the corresponding conversion factor, currently 0.427, 0.662 and 0.043, respectively. The annual effective dose equivalent to be received by the public due to activity in soil and building materials can be calculated by the following formula:

$$\dot{E} = T \times Q \times \dot{D} \times Q_f \times 10^{-6} \qquad (5)$$

where the value of Q is 0.7 Sv/Gy/year for environmental exposure to γ-rays of moderate energy, T is time in hours in one year, i.e., 8760 hours, Q_f is the occupancy factor (0.8), and $\dot{D}$ is the dose rate given in Eq. 4. Thus, the dose rate in air and the annual effective dose equivalent are obtained as 20±2 nGy/hour and 0.10±0.01 mSv/year, respectively. This is within the permissible dose equivalent limit (i.e., 1 mSv/year, ICRP 60, 1990). Similar to Eq. 3, we can also evaluate the external hazard index (H_{ex}) and internal hazard index (H_{in}), which gives the internal exposure to carcinogenic radon and its short-lived progeny, as:

$$H_{ex} = \frac{A_{Th}}{259(\text{Bq/kg})} + \frac{A_{Ra}}{370(\text{Bq/kg})} + \frac{A_K}{4810(\text{Bq/kg})} \qquad (6)$$

$$H_{in} = \frac{A_{Th}}{259(\text{Bq/kg})} + \frac{A_{Ra}}{185(\text{Bq/kg})} + \frac{A_K}{4810(\text{Bq/kg})} \qquad (7)$$

For radon and its short-lived progeny to produce negligible hazardous effects to respiratory organs from materials to be used in construction, both the external and internal hazard index should be less than unity, especially for the value of H_{in}. The obtained values of 0.12±0.02 and 0.15±0.02 for the external and internal hazard index, respectively, are both well below the requirement, indicating that the hazardous effects of this material is negligible.

Radon levels were not measured directly but an upper limit can be inferred from the above guidelines. If the gamma dose rate is lower than 1 mSv/year, then ^{226}Ra concentrations are unlikely to be high enough to cause indoor radon concentrations in excess of the 200 Bq/m^3 guidance level. In this regard, a number of models have been proposed in the literature and checked in this section. These evaluated values are summarized in Table 4, as

Table 4. Radium equivalent activity, dose in air, annual effective dose and the corresponding external and internal hazard indices due to the LUSI mud

Material	Ra_{eq} (Bq/kg)	$\dot{D}$ air (nGy/hour)	Annual effective dose (mSv/year)	Hazard index H_{ex}	Hazard index H_{in}
LUSI mud	43±4	20±2	0.10±0.01	0.12±0.02	0.15±0.02
Faheem *et al.*, 2008	149±40	72.6±42.7	0.30±0.26	0.41±0.27	0.51±0.34
requirement	370	-	1	1	

compared with the mean value of the soil collected from six districts of the Punjab province-Pakistan (Faheem *et al.*, 2008).

4. Conclusion

Analysis was carried out, not only to examine its impact on human health and the environment, but also to explore the potential benefits of the unstoppable hot mud flow by the East Java 'LUSI' Mud Volcano. By examining the entire radionuclide identified in the γ-ray energy spectrum using a large volume and low-background HPGe spectrometer, the re-usability of the mud for brick and cement was ascertained. The results show that the activity concentrations were within the acceptable limits and the use of mud in the construction of buildings is unlikely to give rise to any significant radiation exposure to the occupants. On the whole, the mud samples comply with the parameters outlined in the relevant international legislation and guidance. However, regular monitoring would be advised, due to the fact that two samples exceeded the lower limit of $I \leq 0.5$ which could give rise to doses of 0.3 mSv/year. It is also emphasized that our measured accuracy is improved by a factor of nine at the maximum in virtue of the large volume detector and thorough background subtracted analysis, compared with previous data.

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