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Preliminary Study on Natural Radionuclide Concentration in *Samosir* area

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ABSTRACT

Measurement of natural radionuclides concentration in *Samosir* area of Sumatera island of Indonesia aims to obtain basis data of the content of radionuclides that could be tended in the development of the area. This research was conducted using Neutron Activation Analysis (NAA), which consists of a sampling of soil of Samosir Island as the residential area and the soil surrounding of Pangururan hot-spring as the recreational area which then samples and standards were prepared. Then irradiated with thermal neutron flux 3.5 $\times 10^{13}$ n.cm^{-2}.s^{-1} for 1 hour in a nuclear reactor, and then counted using a gamma spectrometer by HPGe detector. The analysis showed that the soil of *Samosir* Island samples with concentrations of Uranium of (<0.67) ppm and Thorium of (18.00 ± 0.49) ppm and soil surrounding of hot-spring Pangururan detected and found Uranium of (16.83 ± 0. 83) ppm and Thorium of (6.49 ± 0.35) ppm.

Keywords: NAA, Natural radionuclide, Uranium and Thorium, *Samosir* Island.

INTRODUCTION

Radionuclides in the soil come from radioactive dust fall-out, the outcome of weathering of rocks containing natural radionuclides or derived from material from the eruption of the volcano (Artinigsih *et al*, 2008). Radionuclide is atom nucleus with unstable nucleus characterized by excess energy so it will emit radiation; alpha radiation, beta and gamma (Barnes, 1983). Naturally occurring radionuclides are also present in airborne particles as they are also present in soil particles able to be eroded, re-suspended or transported by the processes previously described, and also due to the radon exhalation from soil, which is especially significant to 210pb and 210Po. The dust generated in the manipulation of NORM materials can be another pathway to increase naturally occurring radionuclide content in aerosol particles, although in some cases its contribution to the effective dose

was assessed to be negligible, such as the use of phosphogypsum as soil amendment (Abril *et al*, 2009).

The present of certain number of natural radionuclides in the soil can lead to a danger, whereas soil provides useful resources for the survival of humans and other living creatures. The danger of radionuclides is external or internal radiation. Effect of external radiation is gamma radiation emitted from each radionuclide, while effect of internal radiation is the inhalation of radon and thoron which is gas natural radioactive which product of uranium and thorium decay that affect the incidence of lung cancer (Rasito, *et al*., 2008). Other U-238 decay is Ra-226 is an alpha emitter with a half-life of 1600 year. The origin of this radiation is due to Ra-226 and its decay products, which have been brought up to the earth's surface by the water of hot springs. It enters the body through different paths such as ingestion of food, water or inhalation and replaces calcium and causes cancers and other body disorders due to a long half-life (Aguado *et al*., 2008; Ghiassi-Nejad *et al*,2003; Ushida and Tagami, 2009).

Natural radionuclide can be found in soil (Akpan *et al*, 2016). They did ground investigations of the activity concentrations from primordial radionuclides (238U, 232Th and 40K) in Akpabuyo, Southeastern Nigeria and the result showed the activity concentrations of the radionuclides of 238U, 232Th, and 40K are vary from 2.22 to 116.09 Bq kg^{-1} (with a mean value of 34.67 Bq kg^{-1}), 3.65e87.41 Bq kg^{-1} (mean of 38.59 Bq kg^{-1}) and 6.26e384.99 Bq kg^{-1} (mean of 114.66 Bq kg^{-1}), respectively. Nguelem, et al (2016) reported a research to show activity concentrations in twenty five (25) soil samples collected from various points in bauxite ore deposit in Menoua Division in Western of Cameroon. The measurements of 40K, 226Ra, 232Th, 235U and 238U shown activity of 671 ± 272, 125 ± 58, 157 ± 67, 6 ± 3 and 99 ± 69 Bq kg^{-1}, respectively.

Only a very few studies on radionuclide concentration in Indonesian mountain areas have been found in literature. Sri Artinigsih and Wijiyono (2008) performed a study on the concentration of radionuclides in the soils around the mount of Merapi in Central of Java island of Indonesia. The study conducted after the eruption of the mount Merapi. The concentrations of K-40, Zn-65, Br-82, Zr-97, Sb-121, Ice-137, Bi-211, Pb-212, Bi-214, Pb-214, Sc-216, Th-222, Sb-224, Ra-226, Ac-228, and U-235 were measured. The results showed that natural radionuclides (Ra-226, K-40, Th-222 and U-235) were found in the mount area. It was stated that those natural radionuclides come from lava bursts due to the activities of mount Merapi eruption last date which is usually followed by bursts of material, dust and some rocks. In the past, about 73,000 ± 4,000 years BCE, there was a big eruption of an ancient volcano (Mount Toba) named as Toba eruption. The eruption yielded an island in the middle of its top, known as Samosir Island. This island resulted by the pressure of magma that has not come out of the eruption (Pardosi, 2015). This fact reveals that it is possible that the *Samosir* Island which was the resulted from a volcanic eruption also contain natural radionuclides.

In Northern and Central Sumatra island of Indonesia a wide distribution of ignimbrite and volcanic sediments have been observed. Some geologists considered that these volcanic products were erupted from Lake Toba (Nishimura, *et al.* 1983). In volcanic areas, several hot-springs are usually found. Water of the hot-spring are also found in the areas of Samosir island, one of the hot-spring is Pangururan hot-spring. Water of the hot-spring come from ground-water discharge in the earth's crust after being heated by geothermal and usually temperatures around 37°C (Meinzer, 2002). Because of Samosir Island resulted of a volcanic eruption, the soil of Samosir Island may contain natural radionuclides and also in the soil surrounding of Pangururan hot-pring. And the elements contained in the hot water distributed in the surrounding soil. To the best knowledge of the authors, the research on the natural radionuclides in the soil surrounding of hot-spring of the *Samosir* Island has not found in literature.

This study focuses on determination of concentration and distribution of naturally occurring radionuclides in *Samosir* island of Sumatera Utara province of Indonesia. The objective is to measure the concentration of natural radionuclides in the areas

which suffered from ancient eruption. The result of this study is expected to supply the necessary information for the government of Indonesia.

MATERIALS AND METHODS

Sampling and Sample Preparation

Soil of *Samosir* Island samples were taken from three different locations soil of Samosir Island, which were Tano Ponggol, Kantor Bupati Rianiate and Pasir Putih Parbaba. And soil surrounding of Pangururan hot-spring was taken ± 30 cm from hot-spring pond. Samples were taken in the depth of 5-10 cm from the ground. Soil samples then inserted in polyethylene plastic and sealed. Soil samples were dried by room's temperature. After drying, the samples were crushed and sieved to 200 mesh sizes and then inserted in a polyethylene plastic and sealed.

Washing Polyethylene Vial for Sample Sites

Polyethylene vial inserted into a beaker containing nitric acid (HNO_3 50%), then shaken for 2-3 minutes and soaked for 24 hours. And then dry by room's temperature for 15 minutes. After drying, the vial was inserted into a beaker and sealed to avoid contamination

Irradiation Samples and Counting

Irradiation Soil of Samosir Island Sample

Weighed 0.034740 g of soil of Samosir Island sample and weighed 0.039368 g of standard. Then, inserted each into vials polyethylene (LDPE) and wrapped with aluminum foil. Irradiated with the power is 15 mw and thermal neutron flux $3,5.10^{13}$ $n.cm^{-2}.s^{-1}$ for 1 hour. After irradiation, the samples were allowed to stand for 3-4 weeks.

Irradiation Soil Surrounding of Pangururan Hot-Spring Sample

Weighed 0,039927 g of soil surrounding of Pangururan hot-spring sample and weighed 0,037491 g of standard. Then, inserted each into vials polyethylene (LDPE) and wrapped with aluminum foil. Irradiated with the power is 15 mw and thermal neutron flux $3,5.10^{13}$ $n.cm^{-2}.s^{-1}$ for 1 hour. After irradiation, the samples were allowed to stand for 3-4 weeks.

Counted Using Gamma Spectrometer

Aluminum foil released from each capsules by using tweezers. Then put the activated capsules into HPGe Gamma Spectrometer and closed HPGe detector. Then, counted for 1 hour.

Data Analysis

Qualitative Analysis

The qualitative analysis is based on the characteristics of gamma ray energy for each radionuclides in the sample compared to the energy spectrum of gamma isotope table.

Quantitative Analysis

The quantitative analysis was done by 2 methods, there are the comparative method (comparison) and k_o non-comparative method. The comparative method is conducted by comparing the count of sample with count of standard that

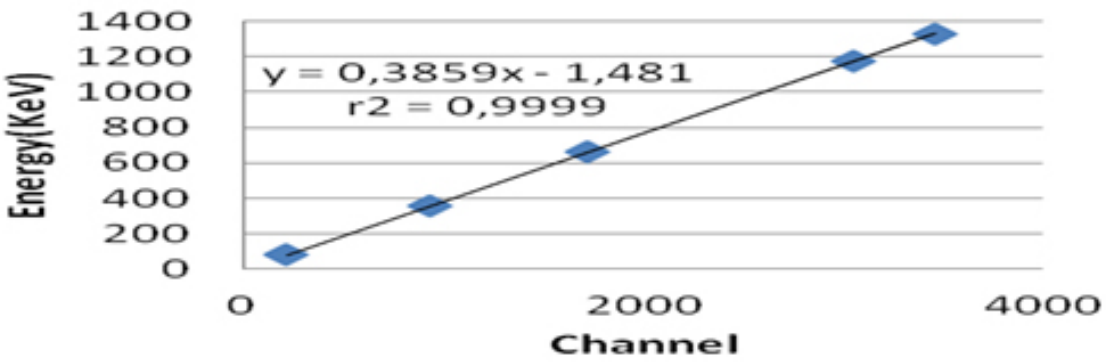

Fig.1: Calibration energy curve

concentration of analyzed elements. Meanwhile, k_o non-comparative method be calculated directly by using k_o –NAA software.

RESULTS AND DISCUSSION

Calibration Curve

By using Ba-133, Cs-137 and Co-60 reference source of gamma Ba-133, Cs-137 and Co-60 calibration data obtained for energy as seen in Table 1, where the number of stripes (Channel) as Xi and energy (keV) as Yi. Determination of the energy curve will affect the quantity of elements in the sample and standard-setting.

By using the data of table 1 obtained a calibration curve energy in the form of linear line made by channel and energy which visible on the monitor screen MCA. As for the energy calibration curve can be seen in Figure 1.

And isotope table of Uranium and Thorium can be seen in Table 2.

Table 1: Data calibration by using standards Ba-133, cs-137 and co-60 source energy

No.	Xi	Yi
1	214	81
2	927	356,8
3	1719	661,66
4	3044	1173,5
5	3457	1332,24

The spectrum of gamma rays that appear in Figure 2 can be seen that the soil of Samosir Island containing natural radionuclides are U-238 (277.60 keV) and Th-232 (312.1 keV). And, soil of Samosir Island also containing elements of Tb-160 (298.7 keV), Yb-169 (307.8 keV), Cr-51 (320.2 keV), La-140 (329.0 keV) and Cs -136 (340.7 keV). High-low peak count is influenced by the amount produced.

Quantitative Determination

Thorium Concentration Determination

Thorium Concentration Determination was done by the comparative method (comparison) conducted by the equation (Mulyaningsih, 2002):

$$W_{sample} = \frac{(Cps)sample}{(Cps)standard} \times w_{standard} \quad ...(1)$$

Where, *W* is weight. Thus, weight of Thorium in soil of Samosir Island sample is:

$$W = \frac{0{,}936389}{0{,}7072} x(0{,}039368\, x\, 12) = 0{,}6254 \text{ mg}$$

With the result that concentration of Thorium in ppm (mg/kg) is:

Table 2 : Isotope table of uranium and thorium

Energy (keV)	Element	Nuclide was Formed	Half-Time ($T^{1/2}$)
312,01	Th	Pa-233	27.0 days
277,60	U	Np-239	2,36 days

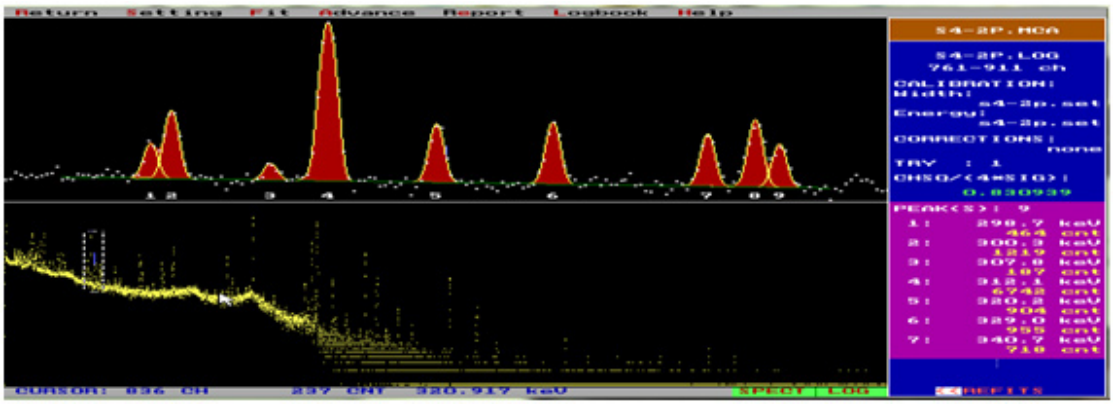

Fig. 2: Gamma energy spectrum of soil of Samosir Island sample

$$[\text{Th-232}] = \frac{0{,}6252}{0{,}034740} = 18{,}00 \text{ mg/kg}$$

Analysis Uncertainty using the equation (Mulyaningsih, 2002) :

$$U = Uc.k \quad ...(2)$$

Uncertainty = concentration of element x U ...(3)

Where U is the uncertainty stretch (%), Uc is the combined uncertainty (%) and *k* is the coverage factor and confidence level of 95%. So we get the uncertainty thorium concentration in soil of Samosir Island sample is:

U = 1,38098244 x 1,96 = 2,706725583 %
Uncertainty = 18,00 x 2,706725583 % = 0,49

Thus, from the above calculations obtained concentration of Th-232 in the soil of Samosir Island is 18.00 ± 0.49 mg / kg.

And, concentration of Thorium in soil surrounding of Pangururan hot-spring sample obtained:

$$W = \frac{0{,}40856}{0{,}7072} \; x \; (0{,}037491 \; x \; 12) = 0{,}2599 \text{ mg}$$

$$[\text{Th-232}] = \frac{0{,}2599}{0{,}039927} = 6{,}49 \text{ mg/kg}$$

By uncertainty,
U = 2,747587 x 1,96 = 5,385271475 %
Uncertainty: 6,49 x 5,385271475 % = 0,35

With the result, concentration of Thorium in soil surrounding of Pangururan hot-spring amounted 6,49±0,35 mg/kg.

Uranium Concentration Determination

Uranium quantitative determination was conducted by k_o non-comparative method. This is because the energy of formation of Uranium (277.60 KeV) only detected in sample, but not in the SRM (Standard Reference Material), so it does not do a comparison between the count data of sample with count data of SRM. The principal parameters of k_o-NAA method has been defined and is already available in k_o-NAA software, so by measuring the amount of counted samples and parameters of the reactor, the concentration of elements in the sample can be calculated directly by k_o-NAA software. By the equation (Corte and Simonits, 1994):

$$\rho_a = \frac{[(Np/t_m)/(S.D.C.W)]_a}{A_{sp.m}} \left(\frac{1}{k_o}\right) \frac{[G_{th.m}.f+G_{e.m}.Q_{o.m}.(\alpha)]\varepsilon_{p.m}}{[G_{th.s}.f+G_{e.a}.Q_{o.a}(\alpha)]\varepsilon_{p.a}} \; 10^6 \quad ...(4)$$

Where, parameters in this equations namely, $\tilde{n}_a$ is the concentration of analyte elements (mg/kg or mg/g); Np is the amount of count is collected at the height of the energy-filled after correction of the missing pulse (the dead time detector and the effect of the coincidence); S is the saturation factor is expressed as $S = 1\text{-}e^{\ddot{e}.tirr}$, ë = decay constancy, ë = (ln 2)/T, with T = radionuclide half-time, t_{irr} = irradiation time (second); D is decay factor = $e^{-\ddot{e}.td}$, t_d = decay time; C is measurement factor = $(1\text{-}e^{-\ddot{e}.tm})/ \ddot{e}.t_m$, t_m = measurement time (second); W is mass of irradiated element (g or kg); $\mathring{a}_p$ is detection efficiency of energy peaks intact including correction for attenuation ã; A_{SP} is a specific count rate, G_{th} is self-absorption correction factor for thermal neutrons; G_e is the self-absorption correction factor for epithermal neutrons; á is parameter for the distribution of neutron flux; $\ddot{o}_e$ ' ÷ $1/E^{1+\acute{a}}$, f is the ratio of the thermal flux of the epithermal; $Q_{\acute{a}}(\acute{a})$ is the ratio between the integral resonance in the cross section of thermal neutrons, and index of a,m are each claiming the analyte and the observe monitor flux;

Factors of k_o involves the principal parameters which can be expressed by equation 5 below:

$$k_o = \frac{[M_m \theta_a \gamma_a \sigma_a]}{[M_a \theta_m \gamma_m \sigma_m]} \quad ...(5)$$

In equation 5, parameters are M = mass number of radioisotope; ã = the fraction of gamma energy emitted by a radioisotope; è = abundance of isotope in nature; ó = cross section of the thermal neutron absorption in the reaction (n, ã).

Therefore, the concentration of uranium in soil of Samosir Island of <0.67 mg/kg, and the concentration of uranium in the soil surrounding of Pangururan hot-spring of 16.83 ± 0.83 mg/kg.

CONCLUSIONS

From the research that has been done obtained the results as:

1. The analysis showed that the soil of Samosir Island samples with concentrations of Uranium of (<0.67) ppm and Thorium of (18.00 ± 0.49) ppm and soil surrounding of hot-spring Pangururan_Samosir detected and found Uranium of (16.83 ± 0. 83) ppm and Thorium of (6.49 ± 0.35) ppm
2. To determine the impact of Thorium and Uranium necessary to measure the concentration of activity emitted by Thorium and Uranium.

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