Research Article

The Influence of Mount Sinabung Volcanic Ash and Phoshate Fertilizers on Natural Radionuclide Content in Agricultural Soils

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Abstract

The concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in four types of chemical fertilizers, the soil samples and Mount Sinabung Volcanic Ash have been measured by gamma spectrometry with a high purity germanium (HpGe) detector. The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K measured for Agricultural Soils are 27,69 Bq/kg, 57,31 Bq/kg, 294,04 Bq/kg (Jeraya Villages) and 34,79 Bq/kg, 84,16 Bq/kg, 175,50 Bq/kg (Tiga Panah Villlages), respectively. Then, activity concentration of ^{226}Ra, ^{232}Th and ^{40}K measured for different fertilizer samples were relatively high for ^{40}K at 3406,605 Bq/kg and relatively low for ^{226}Ra and ^{232}Th at 26,85 Bq/kg and 14,56 Bq/kg, respectively. Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for Volcanic ash are 12,56 Bq/kg, 33,62 Bq/kg and 519,32 Bq/kg, respectively. The radiological hazards of the radium equivalent activity (Raeq), absorbed dose and annual effective are also evaluated in this study.

Keywords: Radionuclide, Fertilizer, Gamma- ray spectrometry, Radium equivalent, Agricultural Soil

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Introduction

Recently, international awareness of NORM as a potential source ionized radiation has increased significantly. Also Naturally Occurring radioactive materials (NORM) became the focus of regulatory interest with the publication of International Atomic Energy Agency and have gained interest among of scientists [1].

Mt. Sinabung, located in Karo District of North Sumatra Province, Indonesia, is a strato volcano having four active craters. Since its latest eruption about 1,200 year ago, a phreatic eruption occurred on August 27th, 2010 and the eruption is still probably occurring until now. Volcanic eruptions result in substantial emissions of gases and ash particles. The areas at risk from volcanic activity encompass a large agricultural productive area.

From the interactions between volcanic ash and human, the danger from volcanic ash containing radioactive nuclides can be categorized into two namely breathing and environmental risks. Breathing volcanic ash may lead to the risk of cancer due to radioelement deposit in the respiratory system. Radionuclides in the environment will expose to the humans in the vicinity [2].

Fertilizers are chemical compounds that provide necessary chemical elements and nutrients to the plants. Fertilizers have becomes essential to the Agricultural field all over the world [3]. Phosphate rocks are the starting material for the production of all phosphate products and main source of phosphorus for fertilizers. Phosphate rock is a general term which refers to rock which is most commonly of apatite group $Ca_5(PO_4)_3$ [F,OH or Cl] [4].

When this rock is processed into the phosphate fertilizers, most radionuclides come into the fertilizers. Thus, fertilizers redistribute naturally occurring radionuclides at trace levels throughout the environment and become a source of radioactivity. This phenomenon may result in potential radiological risks due to possible migration of elements from the agricultural fertilizers to soil and plants, and via food chain, to human begins where this may lead to internal exposure through ingestion of food grown on fertilizer soils [5]. Several studies have shown that phosphate rocks maintain various amounts of naturally occurring radioactive materials (NORM), e.g., uranium, thorium, their decay products and potassium [6-8]. There are two major types of phosphate rocks, (a) sedimentary phosphate, which present about 85% of the phosphate rocks, were formed mainly from organic residues ; they contain 50-200 ppm uranium and 2-20 ppm thorum and (b) ineus phosphate, which supply the remaining parts of phosphate rock, are of volcanic origin; they contain less then 10 ppm uranium but contain an appreciable amount of thorium and rare earths [9].So that the level of activity concentration of radionuclides in phosphate fertilizer provides useful information in the monitoring of environmental contamination.

The aim of this study was to evaluate the Influence of Mount Sinabung Volcanic Ash and Phoshate Fertilizers on Natural Radionuclide Content in Agricultural Soils. Overall, the objective was to determine the radionuclide content in soil plants (chilli, broccoli, corn) with fertilizers and soil no fertilizers, located at Jeraya Village (coordinates 03^{o}08'12.19" N;098^{o}26'10.66" E) and Tiga Panah Village; (coordinates N 03^{o}04.730'; E 098^{o}31.679') where each villages radius of 6-7 km (a high ash fallout area) and 18-19 km (a very low ash fallout area) from Mount Sinabung, Karo Regency, North Sumatra Province, Indonesian. The objective also was to determine Radionuclide Content in fertilizers commonly used in agricultural soil at each villages and volcanic ash of Mount Sinabung.

Materials and Methods

Study Area

The study area are the radius of 6-7 km (namely Jeraya Village; coordinates N 03^{o}08'12.19" and E 098^{o}26'10.66") and 18-19 km (namely Tiga Panah Village; coordinates N 03^{o}04.730' and E 098^{o}31.679') (**Figure 1**) from the Mount Sinabung, Karo Regency, North Sumatra Province, Indonesian Karo District of North Sumatra Province, Indonesia. Both areas of study are also famous for their agricultural activities. So in improving their agricultural productivity, the farmers use fertilizers.

Mount Sinabung is a type B stratro volcano. For the first time, amphreatic explosion occurred on 27 August 2010 and until now eruption is potentially still occur. Ash eruption can cause health problems and ruin farm areas.

Figure 1 Study Area

Sample Collection and Preparation

The soil samples were collected from the radius 6-7 km (namely Jeraya village) and 18-19 km (namely Tiga Panah village) from the Sinabung Mount, North Sumatra, Indonesian. Soil samples were collected from the fields in which different crops and also no crop, four chemical fertilizers types (Nitrogen Phosphorus Potassium (NPK), Triple Super Phospate (TSP), Ammonium Phosphate, Potash Fertilizer (PF)) were collected from the markets and Volcanic Ash were collected from Balcony of resident's house (radius 7-8 km from Mount Sinabung). A simple random procedure was used for soil sampling. Soil samples were collected from the top soil layer (5-20 cm) about 4-5 kg each samples. After Collection, samples were dried and chrushed into a fine powder by using mortar and pestle. Fine quality of the sample was obtained using scientific sieve of 200 micron mesh size. Before measurement, the samples dried in oven at about 105^{o}C for 24 hour to remove the moisture content if any. Samples were palced in Marinelli beaker, or 1 L volume each and weighted. The beakers were sealed using an adhesive to avoid any possibility of Radon leakage. Each samples was stored in a sealed beakers for 30 days to achieve radioactive secular equilibrium when the rate of decay of the daughters becomes equal to that the parent [10].

Measurement of Radionuclide Concentrations with γ-ray Spectroscopy

Radionuclide concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were measured using a HPGe detector manufactured by ORTEC (model: GEM-F5930-XLB-C). It was connected to a personal computer-based data acquisition system, Multi Channel Analyzer model *ORTEC*. Data analysis was carried out via MCA (*Multi Channel analyzer*) *Maestro 7.01*gamma spectroscopy software program. The peak efficiency of the HPGe detector was determined using standard point sources of ^{133}Ba (356.1 keV), ^{137}Cs (661.9 keV) and ^{60}Co (1173.2 and 1332.5 keV). ^{226}Ra activity concentration was estimated from the 609.31 keV γ-peaks of ^{214}Bi or 351.9 keV γ-peaks of ^{214}Pb. ^{232}Th activity concentration was estimated from the 911.1 keV γ-peak of ^{228}Ac. ^{40}K activity concentration was estimated using the 1460 keV γ-peak from ^{40}K itself.

Result and Discussion

Radionuclide Content of the Soil, Volcanic Ash and Fertilizer Samples

The activity concentration of the radionuclides ^{226}Ra, ^{232}Th and^{40}K as well as the corresponding statistical error in the soils, volcanic ash and in the different fertilizer samples under investigation are given in **Table 1**. From the table 1 can be seen that, activity concentration of ^{226}Ra, ^{232}Th and ^{40}K measured for different fertilizer samples were relatively high for ^{40}K at 3406,605 Bq/kg and relatively low for ^{226}Ra and ^{232}Th at 26,85 Bq/kg and 14,56 Bq/kg, respectively. The different and variation of radionuclides concentration in studied chemical fertilizers may be due to the origins of raw material and chemical processing during fertilizer manufacture. Activity concentration ^{226}Ra, ^{232}Th and ^{40}K in soil samples (Tiga Panah Villages) higher than soil samples (Jeraya Villages) because basically the soil control (soil without fertilizers) in Tiga Panah Village was high. According to PP No. 101 (2014) Indonesia, the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in each of samples is well within the permissible limits

Table 1 Activity concentration of Radium, Thorium and Potassium

Sample code	Samples name	Activity Concentration (Bq/kg)		
		^{226}Ra	^{232}Th	^{40}K
Soil 1 (Location : Jeraya Villages, rad 6-7 km from top of Mount Sinabung)				
A1	Soil of Chilli	22,46±2,24	53,88±5,38	265,10±26,09
A2	Soil of Corn	30,65±3,01	63,79±6,30	301,48±29,49
A3	Soill of Brokoli	29,97±2,94	54,25±5,39	315,54±30,75
A4	Soil Control A	23,21±2,31	52,50±5,24	301,98±29,57
Soil 2 (Location : Tiga Panah Villages, rad 18-19 km from top of Mount Sinabung)				
B1	Soil of Chilli	36,92±3,60	94,87±9,23	152,54±15,48
B2	Soil of Corn	35,29±3,47	73,44±7,27	198,89±20,07
B3	Soill of Brokoli	32,16±3,16	84,16±8,25	175,06±17,69
B4	Soil Control B	36,28±3,54	84,48±8,25	150,37±15,31
Fertilizers				
NPK	Nitrogen Phosporus Potassium	11,65±1,21	32,01±3,28	6672,22±627,30
TSP	Triple Super Phosphate	68,55±6,57	8,99±1,13	227,84±22,62
AP	Ammonium Phosphate	0,35±0,11	2,68±0,50	28,86±3,89
PF	Potassium Fertilizer	TTD	TTD	6697,50±629,60
Volcanic Ash of Mount Sinabung				
DV	Volcanic Ash	12,56±1,25	33,62±3,33	519,35±49,41

Radium equivalent activity (Ra_{eq})

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil samples is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of samples containing amounts of ^{226}Ra, ^{232}Th and ^{40}K [11].

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077A_k \qquad (1)$$

Where A_{Ra}, A_{Th}, A_k are the concentrations of the three radionuclides ^{226}Ra, ^{232}Th and ^{40}K respectively, whichis expressed in Bq/kg. Radium equivalent concentration in soils, volcanic ash and different fertilizers are shown in **Table 2.**

The external hazard due to gamma rays corresponds to a maximum radium equivalent activity 370 Bq/kg. This Ra_{eq} is calculated using assumption that 370 Bq/kg ^{226}Ra or 260 Bq/kg ^{232}Th or 4810 Bq/kg ^{40}K produces the same gamma dose rate [12]. The value of Ra_{eq} in each samples must be less than 370 Bq/kg to keep the γ-ray dose below 1.5 mSv y^{-1}. All of the investigated samples still below the recommended worldwide mean value, except NPK and PF because the content of ^{40}K which is the contribution of activity concentration of ^{40}K in NPK and PF higher than the others sample.

Absorbed and Effective dose

The use of phosphate fertilizers in agriculture are further source of possible exposure to public can further be expected in sites being developed for housing [13]. Thus, it is important to calculate the absorbed and annual effective dose, due to the presence of such high levels of radionuclide concentration in the phosphate fertilizers and volcanic ash sample delivered to the worker located in that field.

The measured activity of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses (nGy $h^{-1}Bq^{-1}kg^{-1}$) by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium, respectively [14]

$$D(nGyh^{-1}) = 0.462\ A_{Ra} + 0.604\ A_{Th} + 0.0417\ A_{K} \qquad (2)$$

Where A_{Ra}, A_{Th}, A_k are the concentrations of the three radionuclides ^{226}Ra, ^{232}Th and ^{40}K respectively.

The values of the calculated Absorbed and Effective dose for all samples examined are shown in **Table 2**. The annual effective dose (E) due to γ-rays emitted from radionuclides of ^{226}Ra, ^{232}Th and ^{40}K maintained in the selected samples. To estimate annual effective doses, must be taken into account of the conversion coefficient from absorbed dose in air to effective dose.

Table 2 The Absorbed dose and Annual Effective Dose Outdoor of Soils, Volcanic Ash and Fertilizers

Sample code	Radium equivalent Ra_{eq} (Bq kg^{-1})	Absorbed dose ($nGyh^{-1}$)	Annual Effective Dose Outdoor (mSv/y)
A1	119,92	53,97	0,07
A2	145,08	65,26	0,08
A3	131,84	59,77	0,07
A4	121,54	55,026	0,07
B1	184,33	80,72	0,10
B2	155,62	68,96	0,09
B3	165,99	72,99	0,09
B4	168,67	74,06	0,09
NPK	571,19	302,95	0,37
TSP	98,95	46,60	0,057
AP	6,41	2,98	0,06
PF	515,71	279,29	0,34

$$\text{Outdoor annual effective dose (mSv)} = (\text{Absorbed dose})\ nGyh^{-1} \times 8760h \times 0.2 \times 0.7\ Sv\ Gy^{-1} \times 10^{-6} \qquad (3)$$

Annual estimated average effective dose equivalent received by member is calculated using a conversion factor of 0.7 Sv Gy^{-1}, which is used to convert the absorbed rate to annual effective dose with an outdoor occupancy of 20% for outdoors [15].

According to UNSCEAR that the world wide average limit value for estimation absorbed dose was 59 $nGyh^{-1}$. Based on some of studied samples had absorbed dose greater than the world wide average value and the others still below.

Conclusion

All of the selected samples showed that the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K still below the acceptable limits according to PP No. 101 (2014) Indonesian. Based on the radionuclide results, the radiological hazards of the radium equivalent activities (Raeq) and absorbed dose showed values below than the world-wide safety limit, except NPK and PF which implied that the use of these materials should be under radiation protection.

Acknowledgments

The authors would like to thanks the numerous farmers for the permission to take samples and for information. We also thank PTKMR-BATAN, Indonesia for encouragement and support during the completion of this work.

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Publication History
Received 01st Nov 2017
Revised 24th Nov 2017
Accepted 04th Dec 2017
Online 30th Dec 2017