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Measurements of Natural Radionuclides and ^{137}Cs in Airborne Particulate Samples Collected from Bali and Lombok Islands (Indonesia)

Syarbaini[1*], Wahyudi[2], G. Suhariyono[2], D. Gunawan[3], E. Suharguniyawan[3]

[1]*Center for Radioisotopes and Radiopharmaceuticals Technology, National Nuclear Energy Agency (BATAN), Puspiptek Area Serpong, Tangerang Selatan 15314, Indonesia*

[2]*Center for Radiation Safety Technology and Metrology, National Nuclear Energy Agency (BATAN), Jl. Lebak Bulus Raya No. 49, Jakarta 12440, Indonesia*

[3]*Center for Climate Change and Air Quality, Meteorology Climatology and Geophyisics Agency, Jl. Angkasa I No. 2, Kemayoran Jakarta 10720, Indonesia*

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ABSTRACT

Bali and Lombok islands are popular resorts and tourist destinations in the world because of their culture and beautiful natural view. The natural and anthropogenic radionuclides content in surface air of Bali and Lombok islands such as ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were measured at 3 monitoring stations in a period from January to December 2016. Aerosol samples were collected using a high volume total suspended particles (TSP) sampler. The activity concentrations of those radionuclides in the airborne particulate matter were measured using gamma-ray spectrometry. The results show that the activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K ranged from 1.0 to 3.04 μBq/m^3, not detected to 1.78 μBq/m^3, and 0.03 to 0.49 mBq/m^3, respectively. All the airborne particulate matter filter samples were found to be lower than the minimum detectable activity for ^{137}Cs, which means that none of ^{137}Cs is originated from atmospheric nuclear weapon tests and other sources in the surface air of Bali and Lombok islands. Variations of monthly activity concentrations of natural radionuclides were influenced by rainfall during the study period. Furthermore, the peak concentrations of radionuclides also occur due to volcanic ash coming from the Mount Rinjani eruption.

INTRODUCTION

Radionuclides occurring in atmospheric environment can be classified in two general categories; natural and anthropogenic radionuclides. The main sources of natural radionuclides in the ground level air originate from naturally occurring radioactive including primordial radionuclides ^{40}K and the progeny of the ^{238}U and ^{232}Th decay series and cosmogenic radionuclides produced through interaction of cosmic rays with atmospheric molecules. The existence of natural radionuclides in the ground level air can originate from soil particle resuspension, ash from coal-fired power plants, fossil fuel combustion, phosphate fertilizer and phosphate manufacturing plants, forest fires and volcano eruptions [1,2].

The main global source of anthropogenic radionuclides in the environment is fallout from nuclear weapons tests performed in the atmosphere. Other sources of anthropogenic radionuclides are the nuclear power plants accidents such as Chernobyl and Fukushima, and authorized releases from nuclear fuel processing plants. ^{137}Cs is one of the radionuclides which has been the most intensely used as an indicator of anthropogenic radionuclides contamination in the environment on both regional and worldwide scales. This is because its long physical half-life of 30.2 years, high fission yield, and high bioavailability. Following the Fukushima Dai-ichi Nuclear Power Plant accident, the measurements of ^{137}Cs in the atmosphere have been conducted by several countries [3-5].

*Corresponding author.
E-mail address: sarbaini@batan.go.id
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When reaching the atmosphere, the radionuclides rapidly attach to aerosol particles especially fine particulates, are highly engaged in atmospheric processes. They can be moved over very long distances and can settle at ground level [6,7]. Measurement of natural and anthropogenic radionuclides in ground level air provides important information on the content and origin of the radionuclides for monitoring atmospheric pollution and studying global atmospheric processes [8,9]. The most commonly monitored radionuclides in ground level air are related to their physical and chemical properties such as long physical half-life, readiness to volatilize, and high energy particle emission which can damage tissues and can be accumulated in soft tissue and bones [10]. The occurrence of radionuclides in atmospheric particles has a health impact due to radiological exposure to general public and is considered to be one of the threats for poor air quality in the environments.

Bali and Lombok islands are an absolute tropical paradise and the most popular holiday destination in Indonesia for worldwide travellers and holidaymakers. Many people who come to Indonesia looking for a beach holiday will choose to visit Bali and Lombok. The mountains extend from west to east in the middle of the island of Bali and between these mountains there are clusters of volcanoes, the highest peak of which is Mount Agung as high as 3,142 meters [11].

Lombok Island is located at the east of Bali and it has the shapes and physical characteristics similar to Bali. Lombok island is separated from Bali Island by the Lombok strait and it has a diverse scenic spots to explore on vacation. Mount Rinjani is the highest of the seven mountains exist on the island of Lombok with an altitude of 3,726 m above sea level, and it is one of the highest volcanoes in Indonesia. It has a large caldera with a Crater Lake, 600 m below the rim, and a new volcanic cone which has formed in the center [12].

In Indonesia, information on the concentration of natural and anthropogenic radionuclides in the ambient air is still uncommon. Scientific publication to date on ambient air quality both radionuclides in particulate matter for the recent decade is rare to discover.

The purpose of the present study was to investigate the natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K and anthropogenic ^{137}Cs in the near-surface air collected from the areas Bali and Lombok islands, Indonesia. The study of radionuclides on particulate matter in airborne particulate is expected to establish a scientific foundation for further researches, and it can be a better pollution control to policymakers of the tourist destination areas of Bali and Lombok islands.

METHODOLOGY

Study area and air sampling

Bali island is located between $08^0$03’53”-$08^0$50’48” south latitude and $114^0$25’53”-$115^0$42’40” east longitude with a total area of 5,636.66 square kilometers about 0.29 percent of Indonesia’s total area. Lombok lies between $08^0$33’54” south latitude and $116^0$21’04” east longitude.

Airborne particulate sampling (total suspended particulate) was carried out from January to December, 2016, at Fix Station of Air Quality monitoring, operated by Climate Change and Air Quality Center, Meteorology Climatology and Geophysics Agency, in Bali and Lombok islands, Indonesia. The sampling sites were two locations in Bali island namely Ngurah Rai and Negara with coordinates of 08°44’46” S, 115°09’57” E and 8°20'26.7"S 114°37'03.2"E, respectively, and one location in Lombok island with coordinates of 8°37'58.9"S 116°10'10.8"E, as can be seen in Fig. 1.

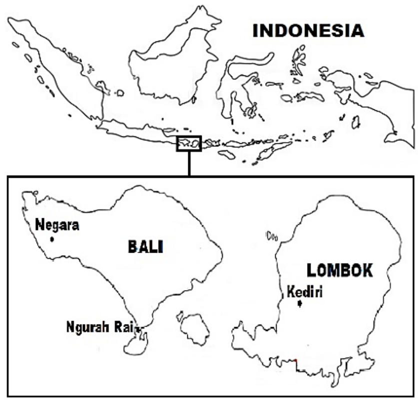

Fig. 1. Map of the study area and sampling sites

A high-volume continuous air sampler (Model TFIA 110-125 V, Staplex, USA) and (Model HV-1000R, Sibata Scientific Technology LTD, Japan) with a rectangular glass fiber filter for TSP (TE-G653 8” x 10”, Tisch Environmental Inc. USA) were used to collect air-borne for 24 h. Airborne particulate samples were collected weekly. The filters were folded in half lengthwise after sampling, so that only surfaces with collected particulate matter were in contact when placed in the filter holder.

The dried filters were weighed before and after airborne particulate sampling to calculate the dust content in the filters by using a microbalance with a sensitivity of 0.0001 mg. Afterwards, filter samples were pressed into pellets to obtain the same geometry as the standard filter using a hydraulic press machine. For the determination of radionuclide activity concentrations, the filter samples were sealed in gas-tight perspect boxes and stored for at least four weeks to allow ingrowth to radioactive equilibrium in the ^{238}U and ^{232}Th series.

Radionuclide analyses

The filter samples were analyzed by a gamma spectrometer using a coaxial HPGe detector with relative detection efficiency of 60 % and the detector shield is made of lead, iron and electrolytical copper. The samples were counted for two to three days. Blank filters were also analyzed and the obtained radionuclide levels were below detection limits. The measurement were performed in underground laboratory with very low level background. The laboratory has ISO/IEC 17025:2017 standard accreditation from the National Accreditation Committe to carry out radioactivity assays in environmental samples and is audited regularly. The ^{226}Ra was measured via the gamma lines of its daughter products ^{214}Pb (295.2, 352.0 keV) and ^{214}Bi (609.3 keV). The ^{232}Th was determined via the gamma lines of the daughter products ^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV) and ^{228}Ac (911.1 keV). The ^{40}K and ^{137}Cs were measured directly from the gamma line of 1460.83 keV and 661.66 keV, respectively.

RESULTS AND DISCUSSION

The variations of monthly average concentrations of Total Suspended Particles (TSP) determined in this study during January to December 2016 are presented in Fig. 2. Monthly average concentration shows that the values vary during different month of the study period, which may be caused by the monthly rainfalls in each sampling site. Dry and rainy seasons in Bali and Lombok islands are influenced by the wind movement coming from Australia (not containing much moisture), and many wind flows containing water vapor come from Asia and the Pacific Ocean. The highest intensity of the rainfall during 2016 occurred in February at Ngurah Rai and Negara stations in Bali island. The lowest number of rainy days at Kediri station in Lombok island was in August and the highest was in November.

A correlation was observed between the mean TSP concentrations and the mean monthly rainfalls. The concentrations of TSP in surface air decrease with the increase of the rainfalls. This is due to the washout of particles from the air by rain, which reduces the concentration of TSP in the airborne particulate material. Some common features were observed in the temporal variations of the rainfalls with minimum values in the rainy weeks and maximum in the dry weeks.

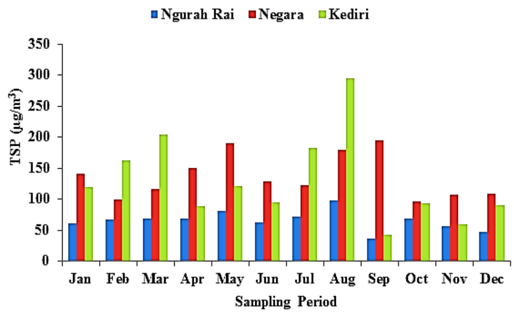

Fig. 2. Distribution of TSP monthly average concentration

During the periods of study, volcano eruptions in Lombok island also significantly influenced mass concentration of paticles. Mount Rinjani, a volcano in the Lombok island, erupted in late October 2015 [13]. Then, a lot of major activities occurred in November 2015 and this eruption has generated ash plumes which significantly influenced the concentrations of paticles until early 2016. In late October 2015, this Mount Rinjani began to erupt causing disruptions in November 2015.

The activity concentrations of radionuclides in air show the trend variation of the monthly TSP measured. Table 1 present the activity concentrations of natural radionuclides and ^{137}Cs determined in this study. The activity concentrations were obtained from weekly values and calculated as an average of 1 month from January to December, 2016. The anthropogenic radionuclide ^{137}Cs activity concentration in all particulates samples were found to be lower than the minimum detectable activity of the gamma counting systems (<MDA). The variations of monthly activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K in air, during January to December 2016 are shown in Fig. 3, 4 and 5.

Table 1. Range of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs contents in air samples

Sampling Site	Coordinates	^{226}Ra µBq/m³	^{232}Th µBq/m³	^{40}K mBq/m³	^{137}Cs µBq/m³
Ngurah Rai	08°44'46" S, 115°09'57" E	1.0 –2.54	n.d. –1.35	0.04–0.12	n.d.
Negara	8°20'26.7"S 114°37'03.2"E	1.37–2.95	n.d. –1.56	0.07–0.49	n.d.
Kediri	8°37'58.9"S 116°10'10.8"E	1.30–3.04	n.d. –1.78	0.03–0.31	n.d.

n.d. : Not detectable (sample analyzed but radionuclide below detection limit)

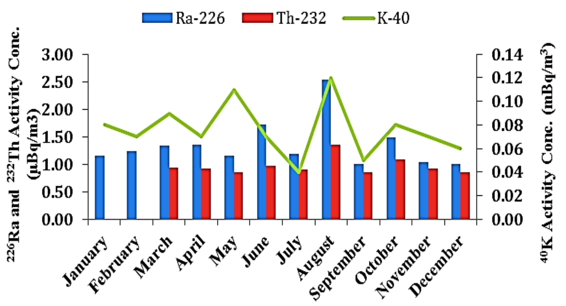

Fig. 3. Distribution of average monthly activities of natural radionuclides, at Ngurah Rai station, Bali island

Some peaks of concentrations of natural radionuclides were observed at Negara and Kediri stations in early 2016, as can be seen in Fig. 4 and 5. However, low concentrations of natural radionuclides were observed at Ngurah Rai station due to the highest intensity of the rainfall during 2016 in February at Ngurah Rai station.

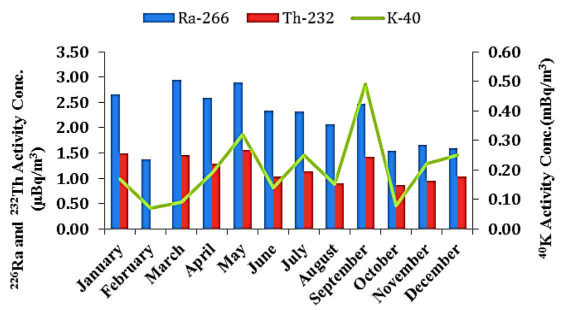

Fig. 4. Distribution of average monthly activities of natural radionuclides, at Negara station, Bali island

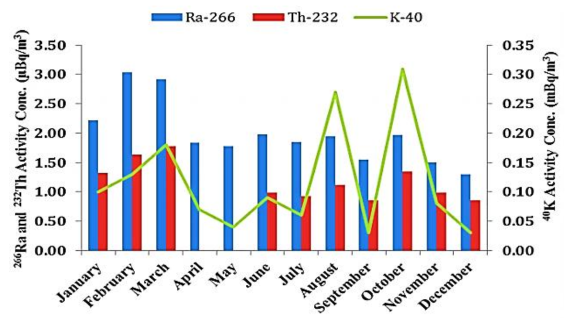

Fig. 5. Distribution of average monthly activities of natural radionuclides, at Kediri Station, Lombok island

A similar event occurred on 1 August of 2016 when Mount Rinjani erupted again. A new eruption that began on 1 August 2016 generated volcanic ash to move at the height 10 km. The volcanic ash particles were mainly fine ash, but there were also coarse ash with particle sizes of 64 µm to 532 µm. There were two directions of volcanic ash particles dispersion, the lower layer was dominated by heading to the east while the upper layer was moving to the west [14]. Injections of *particulate* matter originated from the Mount Rinjani eruption reached Ngurah Rai and Negara stations. The high concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K levels in August, 2016 at Ngurah Rai station and September, 2016 at Negara station were observed as can be seen in Fig. 3 and 4, respectively. In the case of Mount Rinjani's eruption on 1 August 2016 the volcanic ash particles moved away from Kediri station so that there were no significant peak of radionuclides concentration found at this station.

Figures 6, 7 and 8 show the relationship between the concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K and the monthly rainfalls obtained from weekly values in Ngurah Rai, Negara and Kediri stations, respectively. A good correlation was observed between the mean radionuclides concentrations and the mean monthly rainfalls.

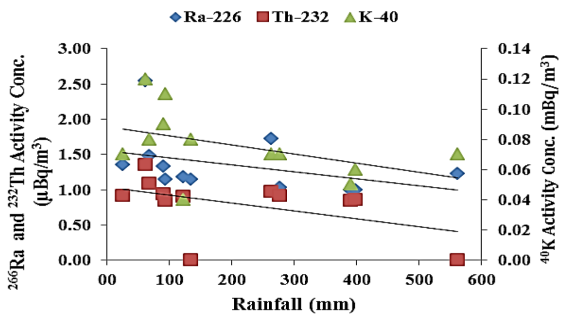

Fig. 6. Relationship between the concentration of radionuclides and rainfalls in Ngurah Rai station, Bali island

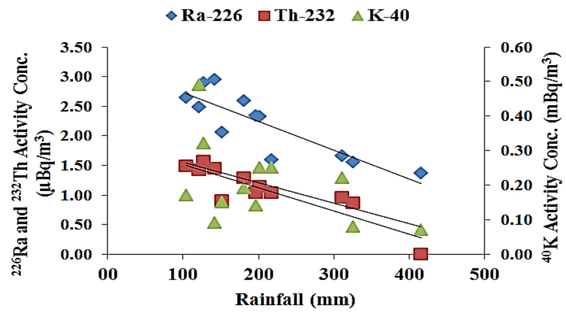

Fig. 7. Relationship between the concentration of radionuclides and rainfalls in Negara station, Bali island

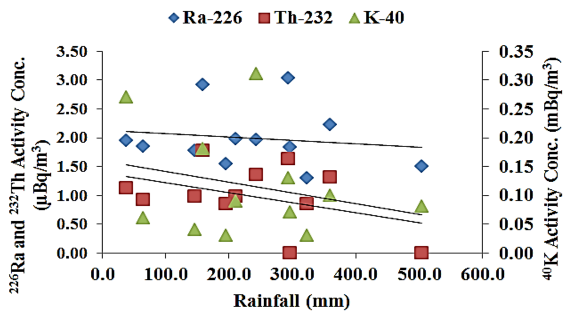

Fig. 8. Relationship between the concentration of radionuclides and rainfalls in Kediri station, Lombok island

In general the average and ranges of activity concentrations of natural radionuclides in air collected from sampling sites at Negara and Kediri stations were higher than that from Ngurah Rai station. Low values of natural radionuclides in air samples collected from Ngurah Rai station, may be correlated with the content of soil particles in ambient air of local area. Ngurah Rai is coastal area that is influenced by the wind coming from marine which does not contain much soil particles resuspension. The source of ^{226}Ra, ^{232}Th and ^{40}K can mainly be attributed to resuspension of soil particles in the lower atmosphere from the local sources of particulate matter at the sampling site.

The measured activity concentrations for ^{226}Ra and ^{232}Th in air samples vary over two orders of magnitude lower than that of ^{40}K. The mean activity concentrations of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K obtained from this study are 1.85 μBq/m^3; 1.11 μBq/m^3; and 0.13 mBq/m^3, respctively. The activity concentration of ^{226}Ra and ^{232}Th are above the world averages values given by United Nations Scientific Committee on the Effects of Atomic Radiation report (UNSCEAR) [15], which are 1.0 and 0.5 μBq/m^3 for ^{226}Ra and ^{232}Th , respectively.

The activity concentration ratio between the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K according to the sampling site are showed in Table 2. The activity concentrations ratios of ^{232}Th/^{226}Ra, ^{226}Ra/^{40}K and ^{232}Th/^{40}K were found to have a wide range with an average value of 0.617; 0.015 and 0.010, which are *almost identically* to other studies reported by Han *et al*. [16]. They measured the natural radionuclides of the atmospheric fine dust (PM_{10}) in Jeju Island Korea and their result showed that the ratios of ^{232}Th/^{238}U, ^{238}U/^{40}K and ^{232}Th/^{40}K were 0.830; 0.037 and 0.031, respectively. It is generally expected that ^{238}U and ^{226}Ra being in the same series are in equilibrium.

Table 2. The activity concentration ratio between 226Ra, 232Th and 40K

Sampling Sites	^{232}Th/^{226}Ra	^{226}Ra/^{40}K	^{232}Th/^{40}K
Ngurah Rai	0.717	0.018	0.013
Negara	0.542	0.011	0.006
Kediri, Lombok	0.591	0.017	0.010
Average	0.617	0.015	0.010

During the period of study, the average of mass concentration were 65.59 μg/m^3, 136.43 μg/m^3, 129.39 μg/m^3 for Ngurah Rai, Negara and Kediri stations, respectiely. Futhermore, the activity concentration of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K were 6.61-36.16 (18.56) Bq/kg-dust, 0 - 23.07 (11.67) Bq/kg-dust, 329.97-3320.60 (1189.1) Bq/kg-dust respctively.

CONCLUSION

The total suspended particles (TSP) collected from surface air in Bali and Lombok islands were measured for the determination of the natural and anthropogenic radionuclides, especially ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs. Except for anthropogenic ^{137}Cs, all the above natural radionuclides were detected in surface air for all sampling sites. The subsequent analysis showed that the distribution of monthly concentrations of radionuclides were influenced by rainfalls and volcanic eruption. The trends of distribution level were found to be maximum during in the dry weeks and minimum values in the rainy weeks. Mount Rinjani eruption, in Lombok island on November 2015 and August 2016 has also given significant peaks concentrations caused by the increasing amount of ash and dust particles thrown into the atmosphere during eruptions.

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AUTHOR CONTRIBUTION

The first author has contributed as the main contributors of this paper. All authors read and approved the final version of the paper.

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