^{210}Pb, ^{235}U, ^{137}Cs, ^{40}K and ^{222}Rn Concentrations in Soil Samples After 2010 Thai and Malaysian Floods

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Abstract. This paper highlights a study of surface soil samples from Northern Malaysia Peninsular (NMP). Soil samples were collected from the topsoil. Concentrations of ^{235}U, ^{210}Pb, ^{40}K and the artificial fallout radionuclide ^{137}Cs in Bq kg^{-1} were determined by γ-ray spectrometry, whereas ^{222}Rn concentrations in Bq m^{-3} were determined by using CR-39 NTDs. ^{210}Pb, ^{235}U and ^{137}Cs concentrations in the soil surface in Penang state were found to be higher than Perlis, Kedah and Perak states, while ^{40}K concentrations in Perak state are higher than Perlis, Kedah and Penang states. Radon concentrations were recorded very high at 306 kBq m^{-3} in Kg. Bukit Sapi, Lenggong in Perak state.

1. Introduction

Study of natural radioactivity is significant to understand the behaviour of natural radionuclides in the soil environment, because such information can be used as the associated parameter values of radiological valuations [1]. Radionuclides concentrations of water and soil are good indicators of the levels of pollution. Thai and Malaysian Floods in 2010, displaced thousands of people is one of the worst natural calamities to hit the country. The flooding in Thailand's south, along a peninsular it shares with Malaysia, has caused chaos in parts of southern Thailand and north Malaysia, making some areas inaccessible. In some places, water levels reached 3 m, and many roads were submersed. In Malaysia, tens of thousands of people have been evacuated in two northern states, Perlis and Kedah. Perlis and Kedah are flooding areas with heavy rainfall so the possible causes of decrease in metals concentration are natural aeration and natural precipitation. Other possible causes of decrease in metal concentration are formation of wetlands, palm oil plantation and the dilution factor of water as it flows downstream. Concentration in topsoil leads to high radionuclide uptake by forest species with shallow root systems (bilberry, mushrooms, and fern). Radon isotopes are members of the natural decay series, that is the ^{238}U decay series (^{222}Rn, $T_{1/2}$=3.8 days), the ^{232}Th decay series (^{220}Rn, $T_{1/2}$=56 s) and the ^{235}U decay series (^{219}Rn, $T_{1/2}$=3.9 s). Almost all radon in the atmosphere is produced in soils and rocks by radioactive decay of the respective precursor, from which it is released and transported to the atmosphere by diffusion. Furthermore, atmospheric ^{222}Rn is the source of its decay products (^{214}Bi, ^{214}Pb, ^{214}Po, ^{210}Pb, ^{210}Bi and ^{210}Po) in the atmosphere. Atmospheric ^{210}Pb is mainly produced within the atmosphere by decay of ^{222}Rn; its direct precursor is ^{214}Po. Previous studies showed that ^{137}Cs distribution in surface soils could be attributed to differences in climatic and topographical situation in a pertinent location [2]. This paper investigates the spatial distributions of ^{210}Pb, ^{235}U, ^{40}k and ^{137}Cs concentrations activity concentrations in the upper ground layer and ^{222}Rn concentrations in subsoil.

1.1 Site description

The study was conducted at NMP, encompasses the four northernmost states on the west coast of Peninsular Malaysia: Perlis, Kedah, Penang and upper portion of North Perak and NMP is geologically divided into the following major regions [3]: Triassic, Quaternary, Tertiary Cambrian, Silurian to Devonian, Carboniferous Intermediate Grad, Ordovician to Silurian, Jurassic to Cretaceous and Permian as shown in fig. 1, and Table 1 shows geographic site of sampling sites. The Malaysia-Thailand border consists of both a land boundary across the Malay Peninsula and maritime boundaries in the Straits of Malacca and the Gulf of Thailand/South China Sea. Malaysia lies to the south of the border while Thailand lies to the north. From west to east, the 646.5 km Malaysia-Thailand border begins at a point which lies just north of the Perlis River where the western most land boundary terminus was to be at "the most seaward point of the northern bank of the estuary of the Perlis River". The state of Penang is a tropical island, just off the north-west coast of Peninsular Malaysia. It is consisted of the island of Penang and a strip of land on the mainland known as Seberang Prai. The state of Kedah is situated in the northwestern corner of Peninsular Malaysia. It covers an area of 9,425 sq. km. Kedah, with the neighboring state Perlis in the north, is traditionally known as the "Rice Bowl of Malaysia", being the major producer of rice in the country. The border town of Bukit Kayu Hitam in northern Kedah is the main gateway to Thailand. Perlis state is bordered by the Thai province of Satun in the north, Kedah in the south-east, and lapped by the gentle waters of the Straits of Malacca on its western coastline. Its land area of 795 sq. km. and population of just 217,480 makes Perlis the smallest state in Malaysia. The annual range of temperature is from 21 °C to 32 °C while the average rainfall is 2000 ml to 2500 ml. Perak lies between Kedah in the north and Selangor to the south.

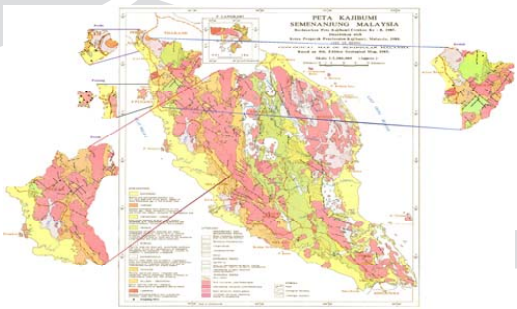

Fig. 1. Geological map of West Malaysia and sampling sites [3]

Table 1. Geographic site of sampling points

SC	Sampling site	N	E
*110	Gelugor, Penang	$05^0$22'20.0"	$100^0$18'56.8"
*404	Alor Relau, Penang	$05^0$20'12.1"	$100^0$16'08.5"
*558	Balik Pulau, Penang	$05^0$20'17.5"	$100^0$11'45.7"
*669	Bukit Dumbar, Penang	$05^0$23'19.1"	$100^0$19'10.7"
*881	Batu Feringghi, Penang	$05^0$28'02.2"	$100^0$15'04.6"
*102	Pantai Aceh, Penang	$05^0$25'19.5"	$100^0$11'49.3"
*111	Penang Hill, Penang	$05^0$25'39.5"	$100^0$16'09.6"
*122	Sg. Planng, Penang	$05^0$23'14.9"	$100^0$12'38.2"
*015	Kg.Pulau Chengai, Kedah	05°49'57.60"	100°29'10.10"
*171	Kg. Rama, Perlis	06°18'18.60"	100°12'23.80"
*201	Kg. Alor Radis, Perlis	06°24'51.00"	100°13'32.70"
*229	Kg. Baru Padang Sanai,Kedah	06°19'58.50"	100°46'36.00"
*259	Kg. Bukit Sapi, Lenggong, Perak	05° 09'3.50"	101°04'17.80"

SC- Sample Code, N- Latitude, E- Longitude

2. Materials and Methods

2.1 Sampling preparation

About 4–6 kg of soil samples were collected from the topsoil ~8 cm using Earth Auger Drill (DZ500). The names of the sampling sites are given in Table 1. The samples were dried at 110 °C-115 °C for 24-28 h in an oven. Dried samples were pulverized and sifted through a 2 mm sieve. Each sample was then weighed and sealed in a 1300 ml Marinelli beaker and analyzed using HPGe detector for 10 h each. They were stored for a period of about 30 days before counting to achieve equilibrium for ^{238}U and ^{232}Th with their respective progeny. The position of the sites was determined using the GPS [4].

2.2 Measurements devices

A gamma ray spectrometry (HPGe) system manufactured by DSG detector systems GmbH, with an energy resolution of 1.85 keV (FWHM) for the 1332.5 keV γ-transition of ^{60}Co, was used to determine the activity concentration of radionuclides ^{210}Pb, ^{235}U, ^{137}Cs and ^{40}K in the samples. The detector was housed inside a massive 5 cm thick old lead shield with 9.5 cm diameter x 20 cm high lead shielding to reduce the gamma ray background, and the analysis of the obtained gamma spectra was performed with the use of software Maestro II (MCA, EG&G ORTEC) with 16384 channels. The activity concentration, in Bq kg^{-1}, of ^{40}K, ^{210}Pb and ^{137}Cs were measured directly from the 1460.81, 46.5 and 661.6 keV, respectively, while activity concentrations of ^{235}U was determined from its γ lines at 143.76, 202.11 and 205.31 keV. The radon level measurements were performed using TASTRAK^{TM} track etch detectors purchased from Track Analysis Systems Ltd (TASL). The passive radon detectors a 2 cm x 2 cm CR-39 were placed at the closed top end of a plastic cup, 6.8 cm in diameter and 7.7 cm length by using a double sided tape, and sponge 1 cm thickness was placed at the another side to a cup to avoid any thoron gas and humidity on detector, a circular hole of diameter 1 cm is made in the center of the lid. The detector chamber is placed inside a PVC pipe 8 cm in diameter and 50 cm in length. The cylinder is buried in the soil at a 50 cm depth and the PVC pipe is open from one end and sealed at the other end to keep water out as shown in Fig. 2. After exposure (26-49 days), the detectors were removed and chemically etched in a 6.25 N, NaOH solution at 70 ^{o}C for 10 h. Alpha particles tracks per cm^2 produced by the decay of ^{222}Rn and its daughters were carried out using an optical microscope with CELLSENS DIGITAL IMAGING software (Olympus, System Microscope BX53) of 100x magnification power as shown in Fig. 2. The observed track density was converted to radon concentration by using the alpha-track density from calibrated CR-39 plastics. The diffusion chamber with detector was calibrated, where a radon calibration sources (3 ^{226}Ra) with 5 μCi were used. The detectors were exposed to 42300 ± 17500 Bq m^{-3} for 7 days by using the portable detector, RAD7 model 711 (Durridge Co. Inc., USA) and the calibration factor = 0.0278 ± 0.00023 track cm^{-2} per Bq d m^{-3} was determined.

Fig. 2. A PVC pipe, an earth drill, and an optical microscope with software used in this study

3. Results and discussion

The concentrations of the radionuclides in the samples of soils are presented in Table 2. ^{210}Pb, ^{235}U, ^{137}Cs and ^{40}K concentrations in the soil surface varies from ND (Pantai Aceh, Penang, Kg. Rama, Perlis, Kg. Baru Padang Sanai, Kedah and Kg. Bukit Sapi, Lenggong, Perak) to 71.56±3.22 Bq kg^{-1} (Gelugor, Penang), ND (Kg.Pulau Chengai, Kedah and Kg. Bukit Sapi, Lenggong, Perak) to 6.42±4.09 Bq kg^{-1} (Batu Feringghi, Penang), ND (Gelugor, Penang, Balik Pulau, Penang, Bukit Dumbar, Penang, Batu Feringghi, Penang and Sg. Planng, Penang) to 0.98±0.49 Bq kg^{-1} (Penang Hill, Penang), and 73.09±11.71 Bq kg^{-1} (Kg.Pulau Chengai, Kedah) to 1334.30 ±32.72 (Kg. Baru Padang Sanai, Kedah), respectively as shown in Fig. 3. ^{210}Pb, ^{235}U and ^{137}Cs concentrations in the soil surface in Penang state higher were found than Perlis, Kedah and Perak states, while ^{40}K concentrations in Perak state are higher than Perlis, Kedah and Penang states.

Table 2. Radionuclides concentrations (Bq kg^{-1}) for sampling sites in NMP

SC	^{210}Pb	^{235}U	^{137}Cs	^{40}K
*110	71.56±3.22	5.44±1.35	ND	1070.83±29.24
*404	10.82±0.96	1.38±0.40	0.83±0.39	326.66±23.33
*558	13.63±1.58	1.49±0.77	ND	607.97±20.42
*669	35.89±2.75	5.09±1.40	ND	999.86±26.90
*881	42.63±2.83	6.42±4.09	ND	897.2±27.89
*102	ND	0.035±0.031	1.04±0.28	191.32±11.80
*111	42.27±3.03	1.45 ±1.54	2.36±1.42	87.68±17.93
*122	11.22±1.19	0.070±0.040	ND	1012.16±27.89
*015	0.13±0.64	ND	0.40±0.35	73.09±11.71
*171	ND	0.39±0.37	0.76±0.39	597.35±23.52
*201	3.43±0.86	0.089±0.039	0.82±035	305.30±17.76
*229	ND	0.021±0.016	0.82±0.19	131.64±7.84
*259	ND	ND	0.57±0.49	1334.30 ±32.72

ND- Not Detected

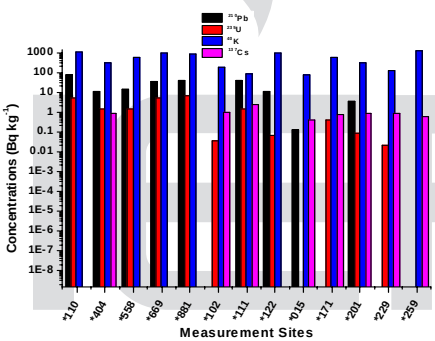

Fig. 3. ^{210}Pb, ^{235}U, ^{137}Cs and ^{40}K concentrations in NMP using HPGe spectroscopy

The soil gas ^{222}Rn concentrations at a 50 cm depth for measurement sites are given in Table 3. In NMP, the higher and lower soil ^{222}Rn concentrations occurred at Kg. Bukit Sapi, Lenggong, Perak and Pantai Aceh, Penang, respectively as shown in Table 3 and Fig. 4. Radon concentrations were recorded very high at 306 kBq m^{-3}, which is exceptionally high as an outdoor value [5]. Tables 4 and 5 show comparisons between ^{210}Pb, ^{235}U, ^{137}Cs, ^{40}K and ^{222}Rn concentrations measurements in soil surface and soil gas with different studies conducted around the globe.

Table 3. ^{222}Rn concentrations for sites in NMP

SC	^{222}Rn (kBq m^{-3})	Standard Deviation	±Standard Error
*110	73.94	6.32	0.80
*404	3.72	0.87	0.20
*558	17.70	2.95	0.37
*669	45.36	5.29	0.67
*881	111.15	6.84	1.37
*102	3.04	0.80	0.14
*111	97.21	8.40	1.31
*122	23.33	2.81	0.51
*015	31.51	3.63	0.68
*171	55.07	5.89	1.15
*201	4.88	1.01	0.19
*229	123.38	7.20	1.55
*259	306.05	32.04	7.14

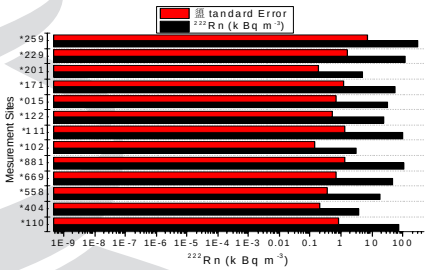

Fig. 4 ^{222}Rn concentrations vs. measurement sites. *259 representative of high concentration, while *102 representative of low concentration in NMP, the scale type Log_{10}.

Table 4. Comparison of gamma ray activity concentrations (Bq kg^{-1}) of surface soil samples with the values reported for other countries of the world.

Countries	Depth (cm)	^{210}Pb	^{235}U	^{137}Cs	^{40}K	References
Serbia	5	45-54	2.6-9.9	29-99	332-925	[6]
France	5-10	……	……	4-11.5	348-802	[7]
Lithuania	5	6.1-158.7	……	1190	215	[8]
Hungary	5-10	……	……	7.6-35.9	256-448	[9]
Jordan	2	……	2.2-6.4	0.9-3.5	179-307.6	[10]
Norway	3	……	……	1-146	31-564	[11]
Malaysia	8	ND-71.56	ND-6.42	ND-2.36	73.09-1334.30	Present study

Table 5. Comparison of ^{222}Rn concentrations (kBq m^{-3}) of soil gas with the values reported for other countries of the world.

Countries	Depth (cm)	^{222}Rn	References
Korea	50	3.9-23.1	[12]
Jordan	50	5-24	[13]
Turkey	50	4.3-9.8	[14]
UK	50	1.30-50.30	[15]
Serbia	50	1.27-155	[16]
Malaysia	50	3.04-306	Present study

4. Conclusions

Concluded that floods and rains reduced radionuclides in soil surface. A ^{222}Rn concentration varies from 3.04 to 306.05 kBq m^{-3}. Radon concentrations were recorded very high at 306 kBq m^{-3} in Kg. Bukit Sapi, Lenggong in Perak state.

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