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Original Article

Natural radioactivity level in fly ash samples and radiological hazard at the landfill area of the coal-fired power plant complex, Vietnam

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ABSTRACT

In this study, natural radioactivity concentrations and dosimetric values of fly ash samples were evaluated for the landfill area of the coal-fired power plant (CFPP) complex at Binh Thuan, Vietnam. The average activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K were 93, 77, 92 and 938 Bq kg^{-1}, respectively. The average results for radon dose, indoor external, internal, and total effective dose equivalent (TEDE) were 5.27, 1.22, 0.16, and 6.65 mSv y^{-1}, respectively. The average emanation fraction for fly ash were 0.028. The excess lifetime cancer risks (ELCR) were recorded as 20.30 $\times 10^{-3}$, 4.26 $\times 10^{-3}$, 0.62 $\times 10^{-3}$, and 25.61 $\times 10^{-3}$ for radon, indoor, outdoor exposures, and total ELCR, respectively. The results indicated that the cover of shielding materials above the landfill area significantly decreased the gamma radiation from the ash and slag in the ascending order: Zeolite < PVC < Soil < Concrete. Total dose of all radionuclides in the landfill site reached its peak at 19.8 years. The obtained data are useful for evaluation of radiation safety when fly ash is used for building material as well as the radiation risk and the overload of the landfill area from operation of these plants for population and workers.

1. Introduction

Environmental radiation background is mostly caused by natural radioisotopes (^{238}U, ^{232}Th decay series, and ^{40}K), and special attention must be paid to the radioactive inert radon gas (^{222}Rn). It was estimated that 50% of the total radiation exposure of the global population is due to ^{222}Rn [1]. Approximately 80% of the total radiation exposure dose to a person per year originates from natural radiation sources [2]. The radiation effect on public health depends on the radiation type, its energy, exposure scenario, geological characteristics, and types of environment [3].

Coal-fired power plants contribute nearly 40.8% of the world's electricity [4]. But they also leave a considerable amount of enhanced mineral matter and radioisotopes that existed in the original coal. Radionuclides are subsequently released into the surrounding area by two pathways: a fraction of nonvolatile nuclides is enriched and concentrated in the ashes, while the volatile other is well known for its polluting potential due to vast releases of various conventional pollutants like CO_2, SO_x, and NO_x into the atmosphere [5].

Bottom ash and fly ash are the leading solid waste of coal combustion in a typical coal-fired power plant (CFPP), and coal burning is the process of enriching some chemical elements, poisonous heavy metals such as lead, arsenic, mercury, and even radioactive elements in the ashes. During the coal combustion, fly ash spreads into the air and is subsequently deposited on the surrounding area of the CFPP. In tandem with high risk as source of particulate air pollution that is currently of great societal concern, fly ash may contain a high amount of radionuclides. Many authors have developed different methods and techniques to determine the radionuclides releases from coal and its burned products at the CFPPs [6–10,11]. [8,11] have found the enrichment of radionuclide contents in fly ash, bottom ash compared with the ones in the feed coals as well as the dependence of the radioactivity concentration in fly ash on the size particle of fly ash [12]. focused on the partitioning behaviour of the radionuclides in the different phases (coal, bottom ash, fly ash). Some studies revealed that the radon exhalation rate from fly ash is less than that from soil and coal, although this fly ash contains a higher concentration of uranium than in the conventional soil [6,7,9]. The minimum surface to volume ratio of

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fly ash is one of the reasons to reduce radon gas escapes from fly ash [13]. [14] applied cover layer and vegetation to reduce the exposure of radon in the waste depository of CFPPs.

[15] reported that 750 million tons of fly ash are discharged by thermal power stations per year worldwide, and it will overgrow in the coming years. In many countries, fly ash is also used as a filler component in building materials, production of ceramic and glass-ceramic, zeolite synthesis [16,17]. However, the amount of fly ash and cinder recycle for diverse purposes is only about 35% of these wastes. The rest has remained in the landfill area, this amount will rise significantly because many new thermal power plants will be put into operation (Viet Nam as an example). It raises a great concern regarding the expansion of the landfill area and the high-level contamination of radionuclides in terrestrial and aquatic environments.

In Vietnam, there are currently 32 CFPPs in operation with the total power of 18,523 MW and 9 others under construction with the total power of 7820 MW. The Vinh Tan 1, Vinh Tan 2 and Vinh Tan 4 CFPPs in Binh Thuan province with the total power of 4244 MW have been put into operation and the Vinh Tan 3 CFPP with the power of 1980 MW will be constructed from 2020 to 2025. In near future, the Vinh Tan CFPPs with the total power of 6224 MW will become the largest CFPP complex in Vietnam. Fly ash and ash discharged during the operation of these coal-fired thermal power plant are transported and stored at the Ho Dua landfill area with an area of about 38.37 ha with a contained volume of about 9.3 million m^3 [18].

Workers who work in this area are constantly and significantly exposed to radiation from fly ash. The radiological risks due to occupational exposure of workers are not only related to the external gamma radiation, but also the internal radiation by fly ash and radon inhalation [19]. Besides, the maximum radiation risk level from total (internal and external) exposure for fly ash should be evaluated if it is dispersed into the surrounding environment or we use the fly ash to produce building materials which human will live in daily. The research of background radiation at the fly ash landfill area aimed at evaluating the effect of background radiation on the workers in the coal-fired thermal power plant area and radiation safety in the utilization of ash and coal slag in land grading and construction.

From the above-mentioned studies, it shows that the amount of ash and slag discharged into the environment is significant. However, the recycling of ash and slag products is still limited. Thus, it will cause certain radiation effects to workers and the surrounding environment of CFPP. Until now there is no full assessment of radiation impacts due to Viet Nam CFPPs operation officially published. In this work, the radiation characterization of fly ash stored in the landfill site of the CFPPs in the Binh Thuan province, Vietnam was studied. The gamma and radon radiation of natural radionuclides in the fly ash samples were measured. Then the radiological effects on local workers and population health were also evaluated. Noted that the total annual effective dose and excess lifetime cancer risks in fly ash samples were determined for both external exposure (by gamma radiation source) and internal exposure (by radon inhalation). Finally, the effects of shielding materials on the slag area with different composition and thickness were evaluated. The results are also preliminary database for our next studies of contamination of radionuclides in soil and water environment surrounding the CFPP and its landfill area.

2. Materials and methods

2.1. Sampling location

Binh Thuan is a province in the middle of Vietnam. It is located in the country's South Central Coast Region with a total area of 7812.8 km^2, a population of 1,576,300 people, and a coastline of 192 km. Binh Thuan's geography is mainly low hills, narrow coastal plains, and narrow terrain along North East - South West.

Vinh Tan CFPP complex, which is located in the Binh Thuan province, has been constructed since 2010 and commissioned in 2018. It involved three CFPPs in operation (Vinh Tan 1, Vinh Tan 2, and Vinh Tan 4). The coal types used in Vinh Tan CFPP complex are domestic anthracite coal for Vinh Tan 1 and Vinh Tan 2 plants; mixture of domestic bituminous coal and bituminous coal from Indonesia and Australia for Vinh Tan 3 plant; bituminous/sub-bituminous coal from Indonesia and Australia for Vinh Tan 4 plant.[1] Fly ash and bottom ash from the operation of these CFPPs are transported to Ho Dua landfill site of an area about 38.37 ha [18]. Fig. 1 illustrated the sampling locations at the fly ash landfill area of Vinh Tan CFPPs.

2.2. Sampling and sample preparation

In present study, the samples were collected at various area of the fly ash landfill area by using a thin steel tube with a diameter of 110 mm and a length of 70 cm. At each sampling location, five fly ash samples of layer (0–30 cm) were collected and mixed to obtain a representative sample. In total, 16 surface fly ash samples were collected, placed in labelled polythene bags, and transported to the laboratory. The samples were then dried at room temperature, crushed to a particle size of less than 0.2 mm. Subsequently, the samples were dried at 105 °C for 24 h and packed into a cylinder container (76.6 mm diameter and height of 20 mm for gamma spectrum analysis and 80 mm diameter and height of 200 mm for radon). Samples then were sealed within 40 days, about 10 half-lives of ^{222}Rn, to reach radioactive equilibrium between ^{226}Ra radionuclide and its descendants. After this period, the samples were measured and analyzed by HPGe spectrometer and Rad 7 detector, respectively.

2.3. Measurement and calculation of activity concentration

2.3.1. Gamma spectrometry analysis

The radioactivity of the samples was measured for 24 h by using the gamma spectrometer with the p-type HPGe detector named GC3520 (Fig. 2). It has a nominal relative efficiency of 35% and the energy resolution of 1.8 keV FWHM at 1332.5 keV energy peak of ^{60}Co. RGU-1, RGTh-1 and RGK-1 reference materials were used in the calibration of the gamma measurement system. Genie 2000 were used for the radioactivity analysis. In which, ^{238}U activity was measured by gamma spectrometry via ^{234}Th (63.38 keV) and ^{234m}Pa (1001 keV). The ^{226}Ra activity was measured by calculate the average of activities of ^{214}Pb (295 keV and 352 keV) and ^{214}Bi (609 keV) in assuming that there are the radioactive equilibrium between ^{226}Ra and its daughters. ^{232}Th activity was measured via ^{212}Pb (238 keV), ^{212}Bi (727 keV) or/and ^{228}Ac (338 keV, 795 keV and 911 keV). ^{40}K activity was estimated by its 1460 keV gamma ray. More details can be found in Refs. [20,21].

2.3.2. Radon analysis – RAD7

The Rad7 radon detector was used to evaluate the activity concentration of ^{222}Rn in the collected fly ash samples with the Grab sample protocol (Fig. 2). Three main operational steps for The RAD7 detector are drying, analysis and cleaning. At first step, the radon gas from the sample was pumped into the detector cell for 5 min, waited for 5 min, and then radon gas was counted for only 5 min. The RAD7 radon detector can detect radioactivity level in a range from 0.37 Bq L^{-1} to 14800 Bq L^{-1}. The ^{222}Rn radionuclide

[1] https://www.sourcewatch.org/index.php/Vinh_Tan_power_station.

Fig. 1. Sampling locations at the fly ash landfill area of Vinh Tan CFPPs.

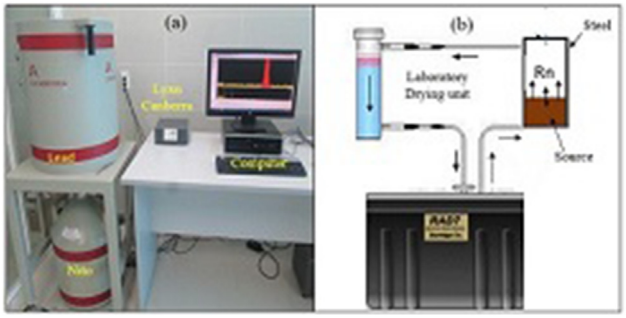

Fig. 2. Experiment layout diagram: (a) HPGe gamma spectrometer; (b) Radon detector – RAD7.

decays by alpha emission to ^{218}Po, and the RAD7 detector calculates the radon concentration activity based on the alpha peak of ^{218}Po. The determination method of radon concentration can be found in Ref. [22].

2.4. Radiation doses and excess lifetime cancer risks

2.4.1. Absorbed dose rates

According to guidelines [23–25], the absorbed dose rates (D) (nGy h^{-1}) at the height of 1 m above the ground are assessed from the gamma radiation of ^{226}Ra, ^{232}Th and ^{40}K natural radionuclides supposed to be uniformly distributed in the ground. They are calculated as follows:

$$D_{in}(\text{nGy h}^{-1}) = 0.92\ A_{Ra} + 1.1\ A_{Th} + 0.081\ A_K \quad (1)$$

$$D_{out}(\text{nGy h}^{-1}) = 0.4368\ A_{Ra} + 0.5993\ A_{Th} + 0.0417\ A_K \quad (2)$$

where, D_{in} and D_{out} are the indoor and outdoor air absorbed dose rates, respectively; A_{Ra}, A_{Th}, A_k are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides (Bq kg^{-1}).

2.4.2. Annual effective doses

The outdoor and indoor annual effective dose E_{out} and E_{in} (mSv y^{-1}) were calculated using the following equations (3) and (4), in which 0.7 Sv Gy^{-1} is a conversion factor from Gy to Sv; 0.2 is outdoor occupancy factor, 0.8 is indoor occupancy factor [20,23–25]:

$$E_{in}(\text{mSv y}^{-1}) = D_{in}(n\text{Gy h}^{-1}) \times 8760(h) \times 0.8 \times 0.7(Sv/Gy) \times 10^{-6} \quad (3)$$

$$E_{out}(\text{mSv y}^{-1}) = D_{out}(n\text{Gy h}^{-1}) \times 8760(h) \times 0.2 \times 0.7(Sv/Gy) \times 10^{-6} \quad (4)$$

The annual effective dose from radon was calculated according to ICRP Publication [26,27]:

$$E_{radon} = (C \times 0.4 \times K \times H) / (3700\ \text{Bq m}^{-3} \times 170\ \text{h}) \quad (5)$$

where E_{radon} is the annual effective dose (mSv y^{-1}), C is the radon concentration (Bq m^{-3}), K is the dose conversion factor (5 mSv WLM^{-1} for occupational workers), H is the annual occupancy at the location, 2160 h for workers (80% of the total time), 170 is the exposure hours taken for WLM [27].

2.4.3. Excess lifetime cancer risks

The excess lifetime cancer risks (ELCR) were calculated according to estimated values of the annual effective doses as expressed in Eqs. (6)–(9) [20,24,25,28]:

$$ELCR_{in} = E_{in} \times DL \times RF \quad (6)$$

$$ELCR_{out} = E_{out} \times DL \times RF \quad (7)$$

$$ELCR_{external} = ELCR_{in} + ELCR_{out} \quad (8)$$

$$ELCR_{internal} = ELCR_{radon} = E_{radon} \times DL \times RF_{radon} \quad (9)$$

where $ELCR_{in}$, $ELCR_{out}$, and $ELCR_{radon}$ are excess lifetime cancer risks for indoor, outdoor, and radon, respectively. DL is the duration of life (70 years), and RF is the risk factor (Sv^{-1}), fatal cancer risk per Sievert. For stochastic effects, ICRP 60 proposed the RF value of 0.05 for the public [29]. RF_{radon} is the risk factor for radon exposure in equilibrium with its progeny. According to ICRP, the value of RF_{radon} is 0.055 Sv^{-1} [28,30]. Total excess lifetime cancer risk was calculated by summing excess lifetime cancer risk for external and internal.

2.4.4. RESRAD-ONSITE

RESRAD-ONSITE code [31] is an extension of the original RESRAD code, RESRAD was developed by Argonne National Laboratory in the 1980s and has been widely used to perform assessments of contaminated sites. The RESRAD-ONSITE computer code evaluates the radiological dose and excess cancer risk to an individual who is exposed while residing and/or working in an area where the soil is contaminated with radionuclides. The modeling of RESRAD-ONSITE considers radiological decay and ingrowth and environmental transport, partitioning, and dilution, governed by the principle of mass conservation over time [31]. In this study, Radiation exposure to workers working directly on landfill was calculated. The issue of radioactive safety for workers working directly and the environment around the fly ash field is taken into use in this study. Calculate the rate of exposure for workers through the replacement of shielding materials and the thickness of materials. The input parameters are given in Table 1.

Table 1
Factors inputs values used in this study.

Parameter	Quantity	UNITS
Area	10000	m^2
Thickness	2	m
Density	1.2	g cm^{-3}
Erosion rate	0.001	m y^{-1}
Total porosity	0.4	
Field capacity	0.2	
Hydraulic conductivity	10	m y^{-1}
Humidity in air	8	g cm^{-3}
Evapostranpiration coefficient	0.5	
Wind speed	2	m s^{-1}
Precipation	1	m y^{-1}
Runoff coefficient	0.2	m y^{-1}
Inhalation rate	8400	m^3 y^{-1}
Mass loading for inhaltion	0.0001	g m^{-3}
Exposure duration	30	y
Indoor dust filtration factor	0.4	
External gamma shielding factor	0.7	
Outdoor time fraction	0.7991	
^{222}Rn emanation coefficient	0.25	
^{220}Rn emanation coefficient	0.15	

3. Results and discussion

3.1. Activity concentrations in the fly ash

The activity concentrations of naturally occurring radionuclides in different fly ash samples were shown in Table 2 and Fig. 3. The average activity concentration of ^{238}U was 93 ± 4 Bq kg^{-1} with a range from 67 ± 8 to 128 ± 12 Bq kg^{-1}; the average value of representative ^{226}Ra was 77 ± 1 Bq kg^{-1} with a range from 68 ± 2 to 91 ± 3 Bq kg^{-1}; the average activity concentration of ^{232}Th was 92 ± 4 Bq kg^{-1} with a range from 58 ± 2 to 129 ± 4 Bq kg^{-1}; for ^{40}K, the activity concentration ranged from 540 ± 34 to 1124 ± 69 Bq kg^{-1} with the average value of 938 ± 30 Bq kg^{-1}. In general, the activity concentrations in fly ash samples were lower than the worldwide average values of 200, 200, 200 Bq kg^{-1} for ^{238}U, ^{226}Ra, ^{232}Th respectively, but two times higher than the worldwide average of 500 Bq kg^{-1} for ^{40}K (following to Ref. [3] for fly ash). In the meanwhile, the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K were 2.66, 2.21, 3.05 and 2.39 times higher than the worldwide average activity of 35, 35, 30 and 400 Bq kg^{-1}, respectively, for soil samples [3], especially the obtained activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides are two times higher than the average activity of 43, 60, 412 Bq kg^{-1} for soil samples in Vietnam [32]. Besides, the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations were higher than the worldwide averages of 50, 50, and 500 Bq kg^{-1}, respectively, for building materials [33]. The average results in this study were found to be a little different from the average world with previous studies. The results show that although the radioactive content in the fly ash sample is lower than the world average, when using fly ash samples in this study we would like to pay attention to the reduction of radioactive content in the sample before they are used for reclamation or construction.

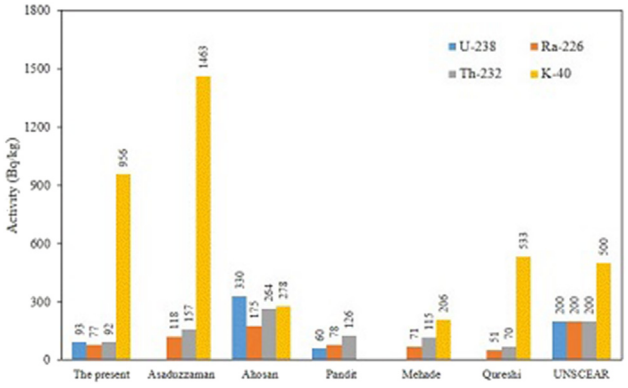

Fig. 3. The average values of activity concentration of fly ash in this study and other countries.

The internal radiological hazards are mostly caused by the inhalation from radon gas (^{222}Rn) - a decayed product with a short half-life of approximately 3.8 days, which is commonly originated during coal combustion at particular temperatures. The results of the radon activity measurement from the fly ash samples were shown in Table 2. Radon activity concentration has been found to vary from 600 ± 20 Bq m^{-3} to 924 ± 38 Bq m^{-3} with an average value of 768 ± 7 Bq m^{-3}. The representative values of the

Table 2
Experimental results of the average activity concentrations in fly ash samples.

No	^{222}Rn (Bq m^{-3})	Activity				Ratio				$E_{mantion}$ (ϵ)
		^{238}U	^{226}Ra	^{232}Th	^{40}K	(1)/(2)	(3)/(1)	(4)/(2)	(4)/(3)	
		(1)	(2)	(3)	(4)					
1	820.9(38.2)	96.1(5.8)	82.9(5.0)	100(6.0)	985.5(59.1)	1.16	1.04	11.88	9.86	0.028
2	760.5(34.1)	77.2(4.6)	78.5(4.7)	93.2(5.6)	986.7(59.2)	0.98	1.21	12.57	10.58	0.031
3	924.4(30.0)	127.9(7.7)	90.6(5.4)	111.1(6.7)	1124.3(67.5)	1.41	0.87	12.41	10.12	0.029
4	600.2(28.6)	87.3(5.2)	68.2(4.1)	81.7(4.9)	862.1(51.7)	1.28	0.94	12.65	10.55	0.028
5	862.1(27.9)	97.1(5.8)	84.9(5.1)	128.5(7.7)	920.7(55.2)	1.14	1.32	10.84	7.17	0.026
6	733.2(27.3)	105.3(6.3)	75.6(4.5)	88.8(5.3)	917.6(55.1)	1.39	0.84	12.14	10.34	0.027
7	734.3(32.7)	67.4(4.0)	73.2(4.4)	88.1(5.3)	936.2(55.2)	0.92	1.31	12.79	10.63	0.028
8	749.8(19.6)	97.7(5.9)	78(4.7)	92.2(5.5)	986.8(59.2)	1.25	0.94	12.66	10.70	0.027
9	613.1(26.2)	76.2(4.6)	73.2(4.4)	88.7(5.3)	933.0(56.0)	1.04	1.17	12.74	10.51	0.023
10	746.4(27.3)	88.2(5.3)	76.3(4.6)	84.0(5.0)	952.7(57.2)	1.16	0.95	12.48	11.34	0.027
11	747.7(30.5)	100(6.0)	71.1(4.3)	86.3(5.3)	919.1(55.1)	1.41	0.86	12.92	10.66	0.029
12	811.4(28.4)	96.2(5.8)	73.3(4.4)	58.1(3.5)	540.2(32.4)	1.31	0.60	11.46	14.46	0.031
13	827.3(22.9)	94.4(5.7)	73.9(4.4)	87.9(5.3)	954.8(57.3)	1.28	0.93	12.92	10.87	0.031
14	732.2(24.0)	91(5.5)	80.4(4.8)	93.0(5.6)	1015.9(61.0)	1.13	1.02	12.63	10.92	0.025
15	895.7(23.7)	110.6(6.6)	81.3(4.9)	96.2(5.8)	1005.1(60.3)	1.36	0.87	12.37	10.44	0.031
16	725.6(29.1)	77.9(4.7)	76.2(4.6)	90.3(5.4)	957.9(57.5)	1.02	1.16	12.56	10.61	0.027
Max	**924.4**(38.2)	**127.9**(7.7)	**90.6**(5.4)	**128.5**(7.7)	**1124.3**(67.5)	**1.41**	**1.32**	**12.92**	**14.46**	**0.031**
Min	**600.2**(19.6)	**67.4**(4.0)	**68.2**(4.1)	**58.1**(3.5)	**540.2**(32.4)	**0.92**	**0.60**	**10.84**	**7.17**	**0.023**
Average	**767.8**(28.2)	**93.2**(5.6)	**77.4**(4.6)	**91.7**(5.5)	**937.4**(56.2)	**1.20**	**1.00**	**12.38**	**10.61**	**0.028**

Note: 820.9(38.2) = 820.9 ± 38.2.

emanation faction for fly ash were varied from 0.031 ± 0.011 to 0.023 ± 0.001 with an average of 0.028 ± 0.001. The values recorded in present was the same as the results reported by Ref. [34] (0.03). The radon emanation fraction for fly ash in the study is lower than for soils (0.2) and rock (0.13) [34]. Because fly ash is a fine grained dust consisting mainly of melted vitreous particles of spherical shape with a smooth surface and it has the minimum surface to volume ratio among all possible particle geometries. The dense glassy structure is considered to be one of the reasons for reducing radon gas escape from fly ash [13], which is consistent with previous radon emanation studies.

For fly ash samples, the average activity of ^{226}Ra, ^{232}Th and ^{40}K in this study are in the range of the obtained values from other studies (in see Fig. 3). In details, in the study of [17]; ^{226}Ra, ^{232}Th and ^{40}K activity concentration of fly ash were 117.8, 157.3 and 1463.3 Bq kg^{-1}, respectively at Bangladeshi, they were 70.9, 115.26 and 205.5 Bq kg^{-1} for samples [35] and 50.1, 70.1 and 533 Bq kg^{-1} for the river sediments of Northern Pakistan [36]. The difference in activity in research works is due to the differences between the origins and number of investigated raw building materials samples. Because, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in fly ash samples depend on their activity concentrations in the coal, the station boiler conditions during the coal combustion of the power, origin and elemental composition in coal [37].

With data of Table 2, the activity concentration of ^{40}K was higher than in comparison with both ^{238}U, ^{226}Ra and ^{232}Th in all fly ash samples of the studied areas. The ratio between the radionuclide concentrations of radioactive isotopes were calculated. The average activity concentrations of ^{232}Th and ^{238}U are similar in the sample. The ratio ^{232}Th/^{238}U varied in the range of 0.6 from 1.32 with an average value of 1.0. The ^{40}K/^{232}Th ratio is a measurement of the correlation between kalium and thorium, its values varied from 7.17 to 14.48 with an average of 10.61. The ^{40}K/^{226}Ra ratios varied from 10.84 to 12.92 with an average of 5.51. In most samples, the comparison was not performed between the ^{238}U activity concentrations from the measurement and from the calculation based on the equilibrium of ^{238}U and ^{226}Ra in the ^{238}U series. The average ^{238}U/^{226}Ra ratios are higher than unity and range from 0.92 to 1.41 with an average of 1.2. The average value of ^{238}U/^{226}Ra) reported by UNSCEAR is about 1.03 [3]. Because, the internal exposure is caused mainly by the inhalation of radon (^{222}Rn) and it is short lived decay products, which is lost during coal combustion at different temperature.

3.2. Assessment of radiological health hazards and excess lifetime cancer risks

The potential radiological hazards to human were assessed by using the estimated activity concentrations for fly ash. The results of absorbed dose rates D_{in} and D_{out}, annual effective doses E_{in} and E_{out}, and excess lifetime cancer risks are shown in Table 3. The values of D_{out} due to gamma radiation generated from ^{226}Ra, ^{232}Th, and ^{40}K varied from 91.47 nGy h^{-1} to 155.81 nGy h^{-1} with an average of 130.24 nGy h^{-1}. The E_{out} values varied from 0.11 to 0.19 mSv y^{-1} with an average of 0.16 mSv y^{-1}. The average gamma dose rate and annual effective dose were higher than the corresponding values of 83.9 nGy h^{-1} and 0.1 mSv y^{-1} in the study of [4]. The excess lifetime cancer risk $ELCR_{out}$ values ranged from 0.43 $\times 10^{-3}$ to 0.74 $\times$ 10^{-3} with an average activity of 0.62 $\times$ 10^{-3}. In the meanwhile, the absorbed dose rate D_{in}, the annual effective dose E_{in}, and the excess lifetime cancer risks $ELCR_{in}$ in the fly ash varied from 175.08 nGy h^{-1} to 296.56 nGy h^{-1} with the average value of 248.02 nGy h^{-1}, from 0.86 mSv y^{-1} to 1.46 mSv y^{-1} with the average value of 1.22 mSv y^{-1}, from 3.01 to 5.10 with the average value of 4.26 respectively. These were slightly lower than the corresponding values reported in previous studies by Ref. [17]. Finally, the annual effective dose and excess lifetime cancer risks of radon varied from 4.12 mSv y^{-1} to 6.35 mSv y^{-1} with an average value of 5.27 mSv y^{-1}, from 15.87 $\times 10^{-3}$ to 24.44 $\times 10^{-3}$ with an average value of 20.3 $\times 10^{-3}$ respectively. The annual effective dose of radon was below the maximum permissible dose limit (10 mSv y^{-1}) for occupational workers recommended by Ref. [26]. However, the excess lifetime cancer risks of radon in Table 3 was about 1.75 times higher than the corresponding values for building materials in the study conducted by Ref. [28]. External annual effective doses and excess lifetime cancer risks for fly ash varied from 0.97 mSv y^{-1} to 1.65 mSv y^{-1} with an average value of 1.38 mSv y^{-1}, and from 3.44 $\times 10^{-3}$ to 5.84 $\times 10^{-3}$ with an average value of 4.88 $\times 10^{-3}$. The results show that when fly ash material is safe in construction and landfilling. However, we need to research regularly and long-term to track the movement of these indices over time.

In comparison with worldwide studies, it was found that the obtained results were higher than the worldwide average values of 59 nGy h^{-1}, 84 nGy h^{-1}, 0.07 mSv y^{-1}, 0.41 mSv y^{-1} for the outdoor, indoor absorbed dose rate, the outdoor, indoor annual effective dose E_{out}, E_{in}, respectively [3] and of 0.29 $\times$ 10^{-3}, 1.16 $\times$ 10^{-3} for the excess lifetime cancer risks $ELCR_{out}$, $ELCR_{in}$ respectively [38]. However, the average outdoor annual effective dose E_{out} of

Table 3
Annual effective dose and excess lifetime cancer risk for fly ash samples.

	E_{radon} (mSvy^{-1})	$ELCR_{radon}$ (10^{-3})	D_{out} (nGyh^{-1})	D_{in} (nGyh^{-1})	E_{out} (mSvy^{-1})	E_{in} (mSvy^{-1})	$E_{external}$ (mSvy^{-1})	$ELCR_{out}$ (10^{-3})	$ELCR_{in}$ (10^{-3})	$ELCR_{external}$ (10^{-3})
1	5.64	21.71	139.77	266.06	0.17	1.31	1.48	0.66	4.57	5.23
2	5.22	20.11	133.74	254.72	0.16	1.25	1.41	0.63	4.38	5.01
3	6.35	24.44	155.81	296.56	0.19	1.46	1.65	0.74	5.10	5.84
4	4.12	15.87	116.78	222.4	0.14	1.09	1.23	0.55	3.82	4.37
5	5.92	22.80	155.25	294.07	0.19	1.44	1.63	0.74	5.05	5.79
6	5.04	19.39	126.78	241.48	0.16	1.19	1.35	0.6	4.15	4.75
7	5.04	19.42	126.07	240.08	0.16	1.18	1.34	0.6	4.13	4.73
8	5.15	19.83	132.85	253.06	0.16	1.24	1.40	0.63	4.35	4.98
9	4.21	16.21	126.34	240.57	0.16	1.18	1.34	0.6	4.13	4.73
10	5.13	19.74	125.73	239.79	0.15	1.18	1.33	0.6	4.12	4.72
11	5.14	19.77	123.28	234.76	0.15	1.15	1.30	0.58	4.03	4.61
12	5.57	21.45	91.47	175.08	0.11	0.86	0.97	0.43	3.01	3.44
13	5.68	21.88	127.03	241.98	0.16	1.19	1.35	0.6	4.16	4.76
14	5.03	19.36	135.68	258.56	0.17	1.27	1.44	0.64	4.44	5.08
15	6.15	23.68	137.58	262.04	0.17	1.29	1.46	0.65	4.50	5.15
16	4.98	19.19	129.7	247.05	0.16	1.21	1.37	0.61	4.25	4.86
Min	**4.12**	**15.87**	**91.47**	**175.08**	**0.11**	**0.86**	**0.97**	**0.43**	**3.01**	**3.44**
Max	**6.35**	**24.44**	**155.81**	**296.56**	**0.19**	**1.46**	**1.65**	**0.74**	**5.10**	**5.84**
Avr	**5.27**	**20.30**	**130.24**	**248.02**	**0.16**	**1.22**	**1.38**	**0.62**	**4.26**	**4.88**

0.16 mSv y^{-1} from this study (Table 3) was in the allowed range from 0.3 to 1 mSv y^{-1} for building materials [39]. It was revealed from these results that the utilization of fly ash as building material did not pose any significant hazards as far as radiation is concerned. Nevertheless, consideration is still necessarily taken into account for recycling fly ash in any aspects of life because of the indoor radiological effects.

Besides, D_{in} values were 1.9 times greater than D_{out} values for fly ash samples. Therefore, when using fly ash materials in buildings, D_{in} consequently became even more critical. The radiation exposure by the radioactivity in building materials could be reduced by either reduction of radon release or gamma-ray protection. In fact, the use of wood, barium element, calcium silicate hydrate, or zeolite can reduce the indoor exposures to levels analogous to outdoor one [40].

To determine the relationship and bond strength between several radiological parameters and radionuclides, Pearson correlation analysis was performed and presented in Table 4. Radon concentrations are strongly correlated with ^{238}U and ^{226}Ra concentrations in the sample, and weakly correlated with ^{232}Th and ^{40}K concentrations. The ^{238}U concentrations are moderately correlated with ^{226}Ra ($r = 0.58$). The ^{238}U concentration has a weak relationship with ^{232}Th and ^{40}K, but ^{226}Ra concentration has a good correlation with ^{232}Th and ^{40}K. It is related to the station boiler conditions during the coal combustion of the power, origin and elemental composition in coal. The correlation value between ^{226}Ra and ^{232}Th is higher than that of ^{226}Ra and ^{40}K. There is a strong correlation between radioactive concentrations of ^{226}Ra, ^{232}Th and ^{40}K with radiation dose and cancer risk ($r > 0.76$). There is no safety margin, and all radiation doses carry some form of risk. Therefore, it is not possible to prevent radiation-induced cancer and lifelong cancer, it can only be minimized by minimizing radiation dose.

3.3. RESRAD-ONSITE

In this study, we used the RESRAD-ONSITE to evaluate the effects of radiation exposure in ash and slag on workers working directly in the landfill site over a period of 30 years. Two circumstances were evaluated: (i) workers working directly in the landfill site without shielding and (ii) workers working directly in the landfill site with shielding.

3.3.1. Workers working daily

Individual and total effective dose rates of ^{40}K, ^{226}Ra, ^{232}Th, and ^{238}U in slag samples in landfill site were calculated and presented in Fig. 4. The results indicated effective dose rates of ^{40}K, ^{226}Ra, ^{232}Th, and ^{238}U on a worker working directly on a landfill site change differently over time. Dose rates of ^{40}K, ^{226}Ra, and ^{238}U gradually decrease, whereas dose rate due to the exposure to ^{232}Th gradually increases over time. For instance, in a period from 0 to 20 years of exposure, dose rates due to the exposure of ^{40}K and ^{226}Ra decrease about 1.78 and 1.06 times. Meanwhile, in the same period, dose rate due to the exposure of ^{232}Th increases about 30.2 times. Total dose rate of all radionuclides in the first exposure year is mostly contributed by ^{40}K and ^{226}Ra, about 51.7% and 45.3%, respectively. Total dose of all radionuclides in landfill site reached its peak at 19.8 years, with ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U contributing doses of 0.105, 0.155, 0.254 and 0.00287 mSv y-1, respectively. The results of Fig. 4 show that during the period from year 20 onwards the total dose rate begins to decrease as the dose rate caused by ^{232}Th slowly increases while the dose induced by K rapidly decreases. Comparing the maximum total dose rate in this study with the value given by Ref. [3]; the results show that the maximum dose rate in this study is higher. Therefore, we need measures to limit radiation exposures (reduce exposure time, use barrier measures) to ensure radiation safety for long-term workers on landfill sites.

3.3.2. Workers working daily with shielding

From calculated results in this study, the radioacitivy concentrations in ash and slag are higher than those in normal soil. It can be explained as the radioactivity enrichment in slag and ash during the operation of coal fired power plant [8,11]. Therefore, scientists and authorities need to evaluate and reduce the radiological impact

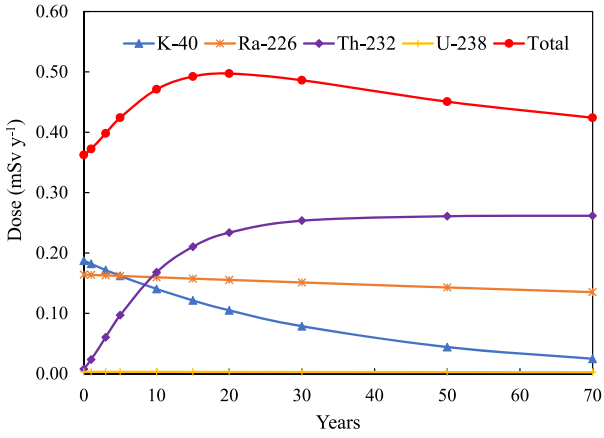

Fig. 4. Total dose due to all radioactivity in fly ash.

Table 4
Pearson's correlation coefficients between radionuclides and related radiological variables.

	^{222}Rn	^{238}U	^{226}Ra	^{232}Th	^{40}K	$E_{manation}$	E_{radon}	$ELCR_{radon}$	D_{out}	D_{in}	E_{out}	E_{in}	$E_{external}$	$ELCR_{out}$	$ELCR_{in}$
^{238}U	0.68														
^{226}Ra	0.75	0.58													
^{232}Th	0.44	0.30	0.74												
^{40}K	0.20	0.21	0.55	0.68											
$E_{manation}$	0.53	0.29	0.00	−0.26	−0.21										
E_{radon}	1.00	0.68	0.75	0.44	0.20	0.53									
$ELCR_{radon}$	1.00	0.68	0.75	0.44	0.20	0.53	1.00								
D_{out}	0.46	0.35	0.81	0.96	0.84	−0.23	0.46	0.46							
D_{in}	0.46	0.36	0.81	0.96	0.85	−0.22	0.47	0.46	1.00						
E_{out}	0.43	0.31	0.76	0.94	0.84	−0.28	0.43	0.43	0.98	0.98					
E_{in}	0.47	0.37	0.81	0.95	0.85	−0.22	0.47	0.47	1.00	1.00	0.98				
$E_{external}$	0.47	0.36	0.81	0.95	0.85	−0.23	0.47	0.47	1.00	1.00	0.99	1.00			
$ELCR_{out}$	0.46	0.35	0.81	0.96	0.84	−0.24	0.46	0.46	1.00	1.00	0.98	1.00	1.00		
$ELCR_{in}$	0.47	0.36	0.81	0.96	0.85	−0.22	0.47	0.47	1.00	1.00	0.98	1.00	1.00	1.00	
$ELCR_{external}$	0.47	0.36	0.81	0.96	0.85	−0.22	0.47	0.47	1.00	1.00	0.98	1.00	1.00	1.00	1.00

Table 5
Total dose with different thickness and shielding material.

Thickness	Total Dose (mSv y^{-1})									
	0	1	3	5	10	15	20	30	50	70
Zeolite (0.7 g cm^{-3})										
0.00	3.63E-02	3.72E-02	3.98E-02	4.24E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.01	3.44E-02	3.57E-02	3.89E-02	4.23E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.05	2.67E-02	2.75E-02	2.97E-02	3.19E-02	3.64E-02	3.92E-02	4.08E-02	4.27E-02	4.51E-02	4.24E-02
0.10	2.05E-02	2.11E-02	2.28E-02	2.45E-02	2.79E-02	2.98E-02	3.09E-02	3.18E-02	3.27E-02	3.47E-02
0.20	1.22E-02	1.26E-02	1.36E-02	1.47E-02	1.67E-02	1.79E-02	1.86E-02	1.90E-02	1.95E-02	2.02E-02
PVC (1.0 g cm^{-3})										
0.00	3.63E-02	3.72E-02	3.98E-02	4.24E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.01	3.02E-02	3.12E-02	3.40E-02	3.69E-02	4.33E-02	4.81E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.05	2.39E-02	2.47E-02	2.67E-02	2.88E-02	3.32E-02	3.60E-02	3.78E-02	4.03E-02	4.51E-02	4.24E-02
0.10	1.65E-02	1.70E-02	1.84E-02	1.99E-02	2.29E-02	2.48E-02	2.60E-02	2.73E-02	2.92E-02	3.21E-02
0.20	7.91E-03	8.17E-03	8.86E-03	9.60E-03	1.11E-02	1.21E-02	1.26E-02	1.33E-02	1.41E-02	1.53E-02
Soil (1.2 g cm^{-3})										
0.00	3.63E-02	3.72E-02	3.98E-02	4.24E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.01	3.25E-02	3.38E-02	3.72E-02	4.09E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.05	2.22E-02	2.29E-02	2.49E-02	2.70E-02	3.13E-02	3.41E-02	3.61E-02	3.89E-02	4.51E-02	4.24E-02
0.10	1.42E-02	1.47E-02	1.60E-02	1.74E-02	2.01E-02	2.20E-02	2.32E-02	2.47E-02	2.72E-02	3.06E-02
0.20	5.92E-03	6.12E-03	6.67E-03	7.25E-03	8.48E-03	9.28E-03	9.80E-03	1.04E-02	1.14E-02	1.28E-02
Concrete (2.3 g cm^{-3})										
0.00	3.63E-02	3.72E-02	3.98E-02	4.24E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.01	2.95E-02	3.09E-02	3.45E-02	3.84E-02	4.71E-02	4.92E-02	4.97E-02	4.86E-02	4.51E-02	4.24E-02
0.05	1.49E-02	1.55E-02	1.71E-02	1.89E-02	2.28E-02	2.58E-02	2.83E-02	3.28E-02	4.51E-02	4.24E-02
0.10	6.44E-03	6.71E-03	7.42E-03	8.20E-03	9.96E-03	1.13E-02	1.24E-02	1.43E-02	1.84E-02	2.40E-02
0.20	1.22E-03	1.27E-03	1.42E-03	1.58E-03	1.96E-03	2.25E-03	2.48E-03	2.86E-03	3.65E-03	4.74E-03

of ash and slag on the surrounding environment. In this study, we evaluated the reduction of total effective dose rate due to the exposure of radioactivity for long-term working workers at the landfill site covered by different shielding materials. The results are presented in Table 5.

In Table 5, when using different shielding materials in the slag area, the results showed that the total doses decrease due to the influence of the composition and thickness of the shielding material. Radiation shielding of materials are in the ascending order: Zeolite < PVC < Soil < Concrete. On the other hand, in the first year, when the shielding materials increase from 0 to 20 cm, total doses from the slag area covered by Zeolite, PVC, Soil and Concrete decease about 2.96, 4.58, 6.12 and 29.76 times. The reason for this reduction is that these shielding will act as absorbent layers of radiation emitted by fly ash in landfills.

The results also indicated that, the total dose received by worker under no shielding in this area is quite large and changes over time: the total dose gradually increased and reached a maximum value of 4.97×10^{-2} mSv/y after 20 years of exposure, then gradually decreased. As the shielding material thickness increases, the absorbed dose decreases and the maximum time is longer. Specifically, when the maximum thickness of about 0.05 m falls within 50 years for the materials being investigated zeolite, PVC, soil, concrete. However, when the shielding thickness is large enough >0.1 m, besides reducing the exposure dose, the maximum dose value exceeds the average life time of humans (70 y). This result shows that among the four materials surveyed, soil has a good shielding effect (better than Zeolite and PVC). Specifically, at a thickness of 0.2 m of soil, the average total dose is reduced by 80%. Soil is a popular, readily available, low-cost material that is suitable for natural ecology. Therefore, the use of soil as a shielding layer for radiation protection is quite effective and appropriate.

4. Conclusion

In this work, the natural radioactivity in fly ash and its radiological hazard to the human health at the landfill site of a Coal Fired Thermal Power Plant complex in Binh Thuan province, Vietnam were evaluated. In general, the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th in the surveyed fly ash samples were lower than the worldwide average values for fly ash, but they were higher than the average world activity for soil and building materials. Radon activity concentrations were higher than the values reported by other researchers for fly ash and building, but the annual effective dose for radon inhalation was below the maximum permissible dose limit (10 mSv y^{-1}) for occupational workers. The radiological hazard indexes for fly ash were higher than the world average values for soil (Some criteria have not given for fly ash). In practice, fly ash material fraction used in building material depends on its product type such as building materials or backfilling materials. Therefore, total annual effective dose due to internal and external radiation exposure emitted from each type product of fly ash building or backfilling materials should be evaluated to ensure the products meet criteria for environmental safety and economically reasonable standard regulations in use. All dose in landfill site reached its peak at 19.8 years, with ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U contributing doses of 0.105, 0.155, 0.254 and 0.00287 mSv y-1, respectively. Using various materials shielding over the slag yard area the results showed that the total dose of radiation in caused was reduced, this deterioration depends on the composition of the shielding material and the thickness of the shielding.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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