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Radiological Consequences of the Use of Fly Ash in Construction Sector and Geotechnical Applications

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Key Words
Cement · Effective dose · Fly ash · Gamma-ray spectrometry · Radiation index

Abstract
This study primarily focuses on the assessment of the radiological consequences for the public from the use of pulverised coal combustion fly ash in construction sector (cement production) and geotechnical applications (embankments, road pavement and road stabilisation) in Turkey. The results of the activity measurements made on samples of the fly ash and Portland cement incorporating 10%, 20% and 30% by mass of fly ash are presented. The results of the external and internal indices, indoor absorbed gamma dose rate and the corresponding annual effective dose were assessed for the samples and these were examined by comparison with the national and international standards or regulation criteria given for the indoor exposure.

Introduction

When lignite coal used in thermal power plants (TPPs) is pulverised and burned inside the boiler coal combustion by-products such as coal slag, bottom ash and fly ash (FA) are obtained. The emission of FA from the stacks into the atmosphere is controlled by emission control devices such as scrubbers, mechanical and electrostatic precipitators. The large fraction of the collected FA is usually stored in piles or dumped in the vicinity of the TPP. In Turkey, existing coal-fired TPPs produce a total of over 15 Mt of FA per year [1]. FA is a hazardous waste because it contains the volatile toxic metals and radionuclides. FA is a very fine powder and tends to travel far in the air. When not properly disposed, it is known to pollute air and water, and causes respiratory problems when inhaled. When it settles on leaves and crops in fields around the TPPs, it lowers the yield. However, its physical and chemical properties make it an ideal raw material for producing high quality and cost-effective building materials such as lightweight concrete, cement, bricks, interlocking pavers, etc. According to coal combustion FA's characteristics potential applications of FA are grouped into the following main categories: construction materials (cement production, concrete, ceramics, glass and glass-ceramics), geotechnical (road pavement and embankment), agriculture (soil amendment) and miscellaneous (sorbent and sludge conditioning) [2]. Each potential application for FA has main advantages such as the use of a zero-cost raw material, the conservation of natural

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sources and the elimination of by-product or waste [3]. Approximately 33% of the total FA produced in Europe is used as cement raw materials, as main constituent in blended cements and as addition for the production of concrete [4], while less than one-hundredth of FA produced in Turkey is used for construction sector (production of cement and concrete). It is known that the incorporation of FA containing enhanced concentration of natural radioactivity in the building materials may affect indoor doses from external irradiation and the inhalation of radon decay products. These building materials are subject to legal control due to the inherent potential risk of exposing members of the public to ionising radiation originated in the material. Therefore, knowledge of the radioactivity levels of materials used in the building industry is important in the assessment of possible radiological hazards to human health. Although several papers have been published about radiological assessment of the use of FA in building materials around the world [5–8], Turkey has few data for the radiometric characterisation of FA from lignite-fired TPPs and the assessment of radiological consequences of the use of FA as a main constituent in the Portland cement production in the construction industry and geotechnical applications [9]. This paper considers the results of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples of FA and Portland cement containing 10%, 20% and 30% by mass FA. In order to assess the radiological consequences of the use of FA in the production of Portland cement and geotechnical applications, external exposure indices such as radium equivalent activity (Ra_{eq}) index, radiation index (RI_1, RI_2 and RI_3) and internal exposure index such as alpha index (I_α), absorbed gamma dose rate (D_R) and corresponding annual effective dose (AED) caused from the external exposure in indoor were evaluated and examined in the light of national and international standards or regulations.

Materials and Methods

Sample Preparation

FA may be siliceous or calcareous in nature. Siliceous FA has pozzolanic properties and is a fine powder of mostly spherical particles. It consists essentially of reactive silicon dioxide (SiO_2) and aluminium oxide (Al_2O_3). The remainder contains iron oxide (Fe_2O_3) and other compounds. Calcareous FA is a fine powder, having hydraulic and/or pozzolanic properties. It consists essentially of reactive calcium oxide (CaO), reactive SiO_2 and Al_2O_3. The remainder contains Fe_2O_3 and other compounds. The proportion of reactive CaO should not be less than 10.0% by mass [10]. The requirements for siliceous and calcareous fly ash for the use as a main constituent of blended cements are defined in TS EN 197-1 [11].

The FA used in this study was obtained from Kangal lignite-fired TTP. Kangal TPP with an installed capacity 457 MW is located in the town of Kangal of the city of Sivas in the eastern Anatolia of Turkey. Kangal TPP consumes approximately 5.2 Mt of lignite coal per year and produces annually 0.8–1 Mt of FA. The total amount of SiO_2, Al_2O_3 and Fe_2O_3 of FA varied in the range of 34–52% while the CaO content varied in the range of 35–38% [12–15]. Portland cement samples contain 70–90% of Portland cement clinker and 10–30% by mass calcareous FA. The samples were dried in a temperature-controlled furnace at 110°C for 10–20 h to remove moisture. After moisture removal, these samples were cooled in moisture-free atmosphere. Each sample was then filled into cylindrical plastic containers, weighed and hermetically sealed. The sealed samples and the reference materials were stored for more than 4 weeks before counting so as to allow ^{224}Ra and ^{226}Ra to reach the secular equilibrium with their short-lived decay products.

Measurement of Activity Concentration

A high-resolution gamma spectrometry system with a p-type coaxial HPGe detector having an active volume of 62 cm^3 connected with standard fast electronics was used to measure the natural radioactivity in the Portland cement samples containing PFA. The detector has a relative efficiency of 12.4% relative to a 7.62 cm × 7.62 cm NaI (Tl) detector, an energy resolution of 1.8 keV at 1332.5 keV of ^{60}Co. The detectors were shielded to reduce gamma-ray background. The absolute efficiency calibration of the gamma spectrometry systems was carried out using a solid nuclide mixture of Amersham gamma reference material containing known activities of ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{137}Cs, ^{88}Y and ^{60}Co. The sample containers were placed on top of the detector for counting. The same geometry was used to determine peak area of samples and references. The background measurements were taken under the same conditions of sample measurements and subtracted in order to get net counts for the sample.

The activity concentrations were averaged from gamma-ray peaks at several energies. The gamma-ray peak of the 351.9 keV from ^{214}Pb and the 609.3 keV from ^{214}Bi were used to determine the activity concentration of ^{226}Ra. The gamma-ray peak of the 911.2 keV from ^{228}Ac

and the 583.2 keV from ^{208}Tl were used to determine the activity concentration of ^{232}Th. The activity concentration of ^{40}K was measured directly by its own gamma-ray peak at 1460.8 keV.

Results

Activity Concentration

The range and mean of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the samples of FA and Portland cement containing 10%, 20% and 30% by mass of FA together with the statistical uncertainty (1σ) are presented in Table 1. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the FA samples were found as $999.6 \pm 74.0\,\mathrm{Bq \cdot kg^{-1}}$ (577.0 ± 25.4 to $1215.0 \pm 47.9\,\mathrm{Bq \cdot kg^{-1}}$), $32.9 \pm 3.0\,\mathrm{Bq \cdot kg^{-1}}$ (22.0 ± 0.8 to $45.0 \pm 1.7\,\mathrm{Bq \cdot kg^{-1}}$) and $224.0 \pm 28.1\,\mathrm{Bq \cdot kg^{-1}}$ (110.0 ± 9.8 to $365.0 \pm 36.9\,\mathrm{Bq \cdot kg^{-1}}$), respectively.

The typical activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured for coal FA samples in the European Union (EU) countries are $180\,\mathrm{Bq \cdot kg^{-1}}$ (maximum $1100\,\mathrm{Bq \cdot kg^{-1}}$), $100\,\mathrm{Bq \cdot kg^{-1}}$ (maximum $300\,\mathrm{Bq \cdot kg^{-1}}$) and $650\,\mathrm{Bq \cdot kg^{-1}}$ (maximum $1500\,\mathrm{Bq \cdot kg^{-1}}$), respectively [16].

As can be seen from Table 1, the mean value of ^{226}Ra was about six times higher than typical value measured in coal FA samples from the EU countries, while the values of the activity concentrations of ^{232}Th and ^{40}K were lower than typical values measured in coal FA samples from the EU countries. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured as 115.3, 16.7 and $160.0\,\mathrm{Bq \cdot kg^{-1}}$ for Portland cement samples with 10% FA; 261.4, 26.1 and $186.6\,\mathrm{Bq \cdot kg^{-1}}$ for Portland cement samples with 20% FA; 447.3, 38.4 and $227.0\,\mathrm{Bq \cdot kg^{-1}}$ for Portland cement samples with 30% FA, respectively. The mean activity concentrations of ^{232}Th and ^{40}K measured in Portland cement samples are compared with the world population-weighted mean values given in UNSCEAR report [17], which are 45 and $420\,\mathrm{Bq \cdot kg^{-1}}$ for ^{232}Th and ^{40}K, respectively. The mean activity concentrations of ^{226}Ra were 3.5–14 times higher than the world population-weighted mean value of $32\,\mathrm{Bq \cdot kg^{-1}}$ for ^{226}Ra.

Discussion

External Exposure Indices

Radium Equivalent Activity Index. A common radiological index, called the radium equivalent activity (Ra_{eq}), has been used to compare the activity concentrations of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K; and to evaluate the radiation hazards associated with these radionuclides. This index is defined based on the fact that $10\,\mathrm{Bq \cdot kg^{-1}}$ of ^{226}Ra, $7\,\mathrm{Bq \cdot kg^{-1}}$ of ^{232}Th and $130\,\mathrm{Bq \cdot kg^{-1}}$ of ^{40}K produce the same gamma dose rate and calculated from the following equation [18]:

$$Ra_{eq} = A_{Ra} + \frac{10}{7} \cdot A_{Th} + \frac{10}{130} \cdot A_K, \qquad (1)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in $\mathrm{Bq \cdot kg^{-1}}$.

The recommended limits of the Ra_{eq} index for building materials are as follows:

Ra_{eq} must be less than $370\,\mathrm{Bq \cdot kg^{-1}}$ for house buildings and less than 740 for $\mathrm{Bq \cdot kg^{-1}}$ for industrial buildings [19].

The range and mean values of the calculated Ra_{eq} for Portland cement samples are shown in Table 2. The mean values of the Ra_{eq} activity index were calculated

Table 1. The range (maximum and minimum) and mean of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the examined samples

Sample		Activity concentration ($\mathrm{Bq \cdot kg^{-1}}$)		
		A_{Ra}	A_{Th}	A_K
Fly ash (FA)	Range	577.0 ± 25.4 to 1215.0 ± 47.9	22.0 ± 0.8 to 45.0 ± 1.7	110.0 ± 9.8 to 365.0 ± 36.9
	Mean ± SE	999.6 ± 74.0	32.9 ± 3.0	224.0 ± 28.1
Portland cement with 10% FA	Range	86.0 ± 6.2 to 134.0 ± 8.2	15.0 ± 0.9 to 18.0 ± 1.1	156.0 ± 15.3 to 164.0 ± 16.1
	Mean ± SE	115.3 ± 14.8	16.7 ± 0.9	160.0 ± 2.3
Portland cement with 20% FA	Range	230.0 ± 12.4 to 283.3 ± 17.6	25.0 ± 1.6 to 27.2 ± 1.7	184.9 ± 18.9 to 189.0 ± 19.1
	Mean ± SE	261.4 ± 16.1	26.1 ± 0.6	186.6 ± 1.2
Portland cement with 30% FA	Range	437.0 ± 31.5 to 458.0 ± 34.8	37.0 ± 2.1 to 40.1 ± 2.3	218.0 ± 21.6 to 234.0 ± 22.9
	Mean ± SE	447.3 ± 6.1	38.4 ± 0.9	227.0 ± 4.7
Fly ash (FA) (EU countries)	Mean	180	100	650
Worldwide mean (UNSCEAR)	Mean	32	45	420

SE: Standard error.

Table 2. The range (maximum and minimum) and mean of the values of radium equivalent (Ra_{eq}) activity index, the radiation index (RI_1), the alpha index (I_α), the indoor absorbed gamma dose rate (D_R) and the corresponding annual effective dose (AED) assessed for the examined cement samples with FA

Sample		Ra_{eq} ($Bq \cdot kg^{-1}$)	RI_1	I_α	D_R	AED
Portland cement with 10% FA	Range	119.4 ± 16.5 to 172.3 ± 18.1	0.41–0.59	0.43–0.67	108.1–156.2	0.53–0.77
	Mean ± SD	151.5 ± 28.2	0.52 ± 0.05	0.58 ± 0.07	137.2 ± 14.8	0.67 ± 0.07
Portland cement with 20% FA	Range	279.9 ± 21.8 to 336.7 ± 25.8	0.95–1.14	1.15–1.42	253.9–305.7	1.25–1.50
	Mean ± SD	313.0 ± 28.2	1.06 ± 0.09	1.31 ± 0.14	284.1 ± 25.9	1.39 ± 0.13
Portland cement with 30% FA	Range	506.6 ± 38.2 to 533.3 ± 41.7	1.71–1.81	2.19–2.29	461.2–484.2	2.26–2.38
	Mean ± SD	519.6 ± 12.7	1.76 ± 0.03	2.24 ± 0.04	471.9 ± 8.2	2.32 ± 0.04

SD: Standard deviation.

as $151.5 \pm 28.2\ Bq \cdot kg^{-1}$, $313.0 \pm 28.2\ Bq \cdot kg^{-1}$ and $519.6 \pm 12.7\ Bq \cdot kg^{-1}$ for Portland cement samples containing 10%, 20% and 30% by mass of FA, respectively. The mean values of the calculated Ra_{eq} for cement samples with 10% and 20% by mass of FA were lower than the recommended limit for house buildings. However, the mean value of the calculated Ra_{eq} for cement samples with 30% by mass of FA was higher than the quoted value, while it was lower than the recommended limit for industrial buildings.

Radiation Index. Radiation index is used only as a screening tool for identifying materials which might be of concern. Any actual decision on restricting the use of a material should be based on a separate dose assessment. Such assessment should be based on scenarios where the material is used in a typical way for the type of material in question. In this study, the radiation index was calculated as proposed by the European Commission report [16]. The Commission suggests that building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at most. Control should be based on dose criterion, which was established considering the overall national circumstances. It is, therefore, recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv. When by-products such as fly ash, bottom ash, slag, phosphogypsum, etc. are incorporated in building materials and if there is sufficient reason to suspect that these contain enhanced levels of natural radionuclides, the activity concentrations of these nuclides in the final product should be measured or assessed reliably from the activities of all component materials. Radiation index (RI_1) is calculated for building materials using the following formula:

$$RI_1 = \frac{A_{Ra}}{300\ Bq\ kg^{-1}} + \frac{A_{Th}}{200\ Bq\ kg^{-1}} + \frac{A_K}{3000\ Bq\ kg^{-1}}, \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in $Bq \cdot kg^{-1}$.

The range and mean values of the RI_1 assessed for the Portland cement samples containing three percentages of FA are presented in the fourth column of Table 2. It is observed in Table 2 that the mean value of RI_1 assessed for Portland cement samples with 10% FA was lower than the criterion of the unity corresponding to an effective dose 1 mSv, while the mean value of RI_1 assessed for Portland cement samples containing with 20% FA was slightly higher than the quoted criterion value. However, the mean value of RI_1 assessed for Portland cement samples containing with 30% FA was 76% higher than the quoted criterion value.

Geotechnical Applications

The recommended limits of the Ra_{eq} index for geotechnical applications are as follows: Ra_{eq} must be less than 2220 for $Bq \cdot kg^{-1}$ for road, railway and bridge constructions (roadbeds and road pavement) and less than 3700 for $Bq \cdot kg^{-1}$ for utilisation in foundation of non-residential buildings (structural fill and embankments) [19].

The mean value of the Ra_{eq} activity index was calculated as $1063.8 \pm 217.6\ Bq \cdot kg^{-1}$ (616.9 ± 32.6 to $1307.4 \pm 53.6\ Bq \cdot kg^{-1}$) for FA samples. The mean value of the Ra_{eq} was lower than the recommended limits of 2220 and 3700 for $Bq \cdot kg^{-1}$.

The radiation index (RI_2) was calculated for materials used in road, street and related construction work using the formula [20]:

$$RI_2 = \frac{A_{Ra}}{700\ Bq \cdot kg^{-1}} + \frac{A_{Th}}{500\ Bq \cdot kg^{-1}} + \frac{A_K}{8000\ Bq \cdot kg^{-1}}, \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in $Bq \cdot kg^{-1}$.

The radiation index (RI_3) was calculated for materials used in landfill and landscaping using the formula [20]:

$$RI_3 = \frac{A_{Ra}}{2000\ Bq \cdot kg^{-1}} + \frac{A_{Th}}{1500\ Bq \cdot kg^{-1}} + \frac{A_K}{20000\ Bq \cdot kg^{-1}}, \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in $Bq \cdot kg^{-1}$.

The activity indices must be less than unity for materials used in road, street roadbeds, road pavement, landfill, landscaping, structural fill, embankments, etc.

The mean values of RI_2 and RI_3 were calculated as 1.52 ± 0.31 (0.88–1.87) and 0.53 ± 0.11 (0.31–0.66) for the examined FA samples. It can be seen that the mean value of RI_2 was 52% higher than the unity, while the mean value of RI_3 was 47% lower than the unity.

Internal Exposure Index (Alpha Index)

Alpha index was developed for the assessment of the excess alpha radiation due to inhalation originating from building materials. The alpha index (I_α) was determined by the next formula [21]:

$$I_\alpha = \frac{A_{Ra}}{200\ Bq \cdot kg^{-1}}, \quad (5)$$

where A_{Ra} is the activity concentration of ^{226}Ra in $Bq \cdot kg^{-1}$.

The design level for indoor radon exposure in future constructions set by Turkish Standard TS 12614 [22] corresponds to an annual mean radon gas concentration of $200\ Bq \cdot m^{-3}$, while recommended value for remediation of indoor radon exposure in existing building set by Turkish Radiation Safety Regulation is $400\ Bq \cdot m^{-3}$. $I_\alpha \leq 1$ corresponds to the ^{226}Ra activity concentration less than or equal $200\ Bq \cdot kg^{-1}$. When the activity concentration of ^{226}Ra in a building material exceeds the value of $200\ Bq \cdot kg^{-1}$, it is possible that the radon exhalation from this material could cause indoor radon concentration exceeding $200\ Bq \cdot m^{-3}$. The range and mean values of the calculated I_α for the examined Portland cement samples, are presented in the fifth column of Table 2. It is observed that the mean value of I_α assessed for Portland cement samples with 10% FA was lower than the recommended design level, while the mean values of I_α assessed for cement samples with 20% and 30% FA were higher than the quoted level.

Indoor Absorbed Gamma Dose Rate

The absorbed dose rate in indoor air, due to gamma ray emission from the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the building material, was assessed using data and formula provided by the EC report [16]. The indoor absorbed gamma dose rate (D_R) was estimated from the following equation:

$$D_R\left(nGy \cdot h^{-1}\right) = 0.92 \times A_{Ra} + 1.1 \times A_{Th} + 0.08 \times A_K, \quad (6)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in $Bq \cdot kg^{-1}$. The estimated results for the D_R are given in the sixth column of Table 2.

From the data in Table 2, the estimated mean values of D_R were 1.6–5.6 times higher than the world mean (populated-weighted) indoor absorbed gamma dose rate of $84\ nGy \cdot h^{-1}$.

Annual Effective Dose

To estimate the effective dose rate, one has to take into account the conversion factor from absorbed dose in air to effective dose and the indoor occupancy factor. In the recent UNSCEAR report [17], a value of $0.7\ Sv \cdot Gy^{-1}$ was used for the conversion factor from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 80% of time is spent indoors, on average, around the world. The annual effective dose (AED) in units of mSv per year was estimated using the following formula:

$$AED = D_R\left(nGy \cdot h^{-1}\right) \times 8766\ h \cdot y^{-1} \times 0.8 \times 0.7\ Sv \cdot Gy^{-1} \times 10^{-6}, \quad (7)$$

where D_R ($nGy\ h^{-1}$) is given by Equation (6).

The estimated results for AED are given in the seventh column of Table 2. The mean values of the AED estimated for cement samples with 10% FA was lower than the annual effective dose criterion of 1 mSv, while the mean values of AED assessed for cement samples with 20% and 30% FA were higher than the quoted dose criterion value.

Conclusions

Significant economic and environmental problems concerning the handling and disposal of coal FA have led to the implementation of various alternative utilisations. In Turkey, lignite coal combustion FA has been utilised as building materials and/or building material additives in cement and concrete sectors. Restrictions or recommendations concerning the activity concentrations of natural radionuclides in materials used in house and

industrial building and geotechnical applications, and the radon concentration in air of homes, schools and workplaces have been put into practice to avoid significant overdoses to exposed residents in the European Union (EU) and non-EU countries. Turkey applies a strict criterion based on a dose in the range 0.3–1 $mSv \cdot y^{-1}$ for building materials used in bulk amounts, for example concrete. The design level for indoor radon exposure in homes and schools was set by the Turkish Standard TS 12614.

In this study, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and external and internal exposure indices, the indoor absorbed gamma dose rate and the corresponding annual effective dose have been determined to assess the radiological consequences of the use of lignite coal FA from Kangal TPP for production of Portland cement and geotechnical applications in Turkey. The results showed that the mean values of the Ra_{eq}, RI_1, I_α and AED assessed for Portland cement samples containing 10% by mass of FA were below the recommend limit or the recommended design level. However, the mean values of RI_1, I_α and AED assessed for Portland cement samples containing 20% and 30% by mass of FA were above the recommended design level. As a result, the examined Portland cement products with 10% FA comply with the national and international regulations, setting limits on the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in building products. Also, the values obtained for FA samples collected from Kangal TPP were within the recommended safety limit for geotechnical applications.

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