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Establishing a Relationship between Coal Quality and the Enrichment of Radionuclides in Coal Combustion Residues

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Abstract

Coal-fired power plants (CFPP) provide approximately 40% of the world's energy demand. Naturally occurring radioactive materials (NORM) contained in coal become enriched in coal combustion residues as a result of the elimination of carbon during combustion. The fly ash and bottom ash produced from CFPP may be significant sources of exposure to naturally occurring radionuclides for the population near the combustion plant or ash dumps. Despite this fact, very few studies have actually addressed the relationship of the NORM enrichment factors and the quality of coal used. This paper aims to relate the quality of coal to the enrichment factors for the radionuclides of interest (K^{40}, Ra^{226}, Th^{232} and Po^{210}) in coal combustion residues from three South African CFPP. The data from other CFPP was also taken into account to establish this correlation. The feedstock coal used in these CFPP is typically low quality, with ash content in the range of 25 - 45 wt%. The radionuclides investigated were determined by gamma spectrometry with the exception of Po^{210}, which was determined by alpha spectrometry. The enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash and bottom ash (except Po^{210}) was found to be directly proportional to the quality of coal. That is when the ash percentage increased (coal quality decreased) the enrichment factor decreased. The Po^{210} radionuclide in the bottom ash had an enrichment factor less than one. The relationship between coal quality and enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} in both the fly ash and bottom ash (except Po^{210} in the bottom ash) was demonstrated by the following mathematical equation:

$\text{Enrichment Factor} = \dfrac{1}{\text{Ash}(\%)\text{ in feed coal}}$. This equation may be used as a good indication in obtaining an estimate in determining the enrichment of the mentioned radionuclides in coal combustion products such as fly ash and bottom ash.

Keywords

Radionuclides, Coal, Enrichment Factor, Coal Quality

1. Introduction

Coal-fired power plants (CFPP) provide approximately 40% of the world's energy demand [1]. In developing countries, such as South Africa, coal has a significant role in power generation and contributes to approximately 77% of the country's energy [2]. During coal combustion, the elements in minerals and organic fractions of coal are liberated and distributed into combustion products *i.e.* fly ash, bottom ash and flue gas [3]. The increase in thermal generation capacity and subsequently a deterioration of the quality of coal used has resulted in increased generation of fly ash and bottom ash of varying properties. Naturally occurring radioactive materials (NORM) are among the inorganic constituents that are present in coal. These radioactive materials are enriched in coal combustion products such as fly ash and bottom ash following the combustion of coal [4]. The installation of ash collectors like cyclones, electrostatic separators and bag filters significantly reduces the emission of radionuclides to the atmosphere [5]. In contrast, the treatment and disposal of power plant ash continues to be problematic, particularly in South Africa where the coal has an inherent high ash content (low quality), typically between 25% - 50%, and thus a significant amount of ash is generated.

The interest in measuring NORM concentrations in coal and resulting combustion residues (such as fly ash and bottom ash) is due to the awareness of health hazards and environmental pollution [6]. The fly ash and bottom ash produced from CFPP are significant sources of exposure for the population near the plant to naturally occurring radionuclides [7] [8]. The naturally occurring radionuclides, particularly K^{40}, Ra^{226}, Th^{232} and Po^{210} released by these plants pose a potential health hazard [9]. Due to its short half-life and the highly energetic cell-disrupting alpha particles (5.3 MeV) emitted during its decay process, Po^{210} is considered a major health hazard [10]. Once Po^{210} from the air is assimilated into flora and fauna, it may be bio-concentrated in the food chain and, thus, poses further threat to human health [11].

The concentration of most radioactive elements in solid combustion wastes, such as fly ash and bottom ash, will be multiple times higher than the concentration in the original coal [12]. The enrichment factors of certain radionuclides can sometimes be a few folds or even several magnitudes, relative to the feed coals [13]. The number of natural radionuclides discharged into the atmosphere via the ash produced from a CFPP depends on the ash content of coal, the temperature of combustion, partitioning between fly ash and bottom ash, and the efficiency of control devices [5] [14].

Several studies wherein coal enrichment factors with respect to certain radionuclides in coal and coal combustion products were conducted by other authors [4] [5] [10] [15]. However, none of these studies actually relates the quality of coal to the enrichment of radionuclides (enrichment factors) in the fly ash and bottom ash. This attribute of coal quality is one that every CFPP is aware of in terms of the coal being burnt. Therefore, knowing the relationship between coal quality and enrichment factors may enable us to determine the concentration of radionuclides in the combustion products prior to the combustion of coal. This may prevent scenarios such as those in China whereby it was found that some coal ash was too radioactive for reuse in building materials [16]. For this very reason, radioactivity in coal and coal combustion residues' uses are being limited [17].

It is hypothesized that the relationship between the concentration of radionuclides in coal and the combustion products *i.e.* fly ash and bottom ash, is closely related to the quality of coal. Hence, the objectives of this study are to evaluate the ash percentage content (hence quality) of 3 different coals fed to 3 different CFPP and evaluate the enrichment factors in the fly ash and bottom ash (in comparison to other studies as well). As a result, thereof, a mathematical correlation between coal quality and the enrichment factor of radionuclides in coal (in terms of an equation) is to be established, since it does not exist in present-day literature.

2. Materials and Methods

2.1. Sample Collection

The feed coal to 3 different CFPP from 3 different coalfields in South Africa was used in this study. The coal mines (**Figure 1**) are in close proximity to the CFPP. Monthly composite samples were sampled on alternate days over a period of 3 months. The three-month period of sampling assured that the samples were representative of the feed coal to each CFPP.

The proximate and ultimate analysis for the feed coal provided for these samples by the power supplier indicated that, over the three months, the feed to the plant was consistent. On the same days as the feed coal was sampled, the fly ash (from the hoppers) and bottom ash (from the boiler) from the 3 CFPP were also sampled. This ensured that the fly ash and bottom ash were indeed products of the sampled feed coal. The gross samples were then air-dried, milled (coal and bottom ash) and carefully split in accordance with ISO recommendations in order to obtain a representative sample of particle size < 250 um prior to chemical analyses.

2.2. Sample Processing and Analyses

The feed coal samples were supplied together with the proximate data by the power utility and were analysed in accordance with ISO 18283:2006 [19] and

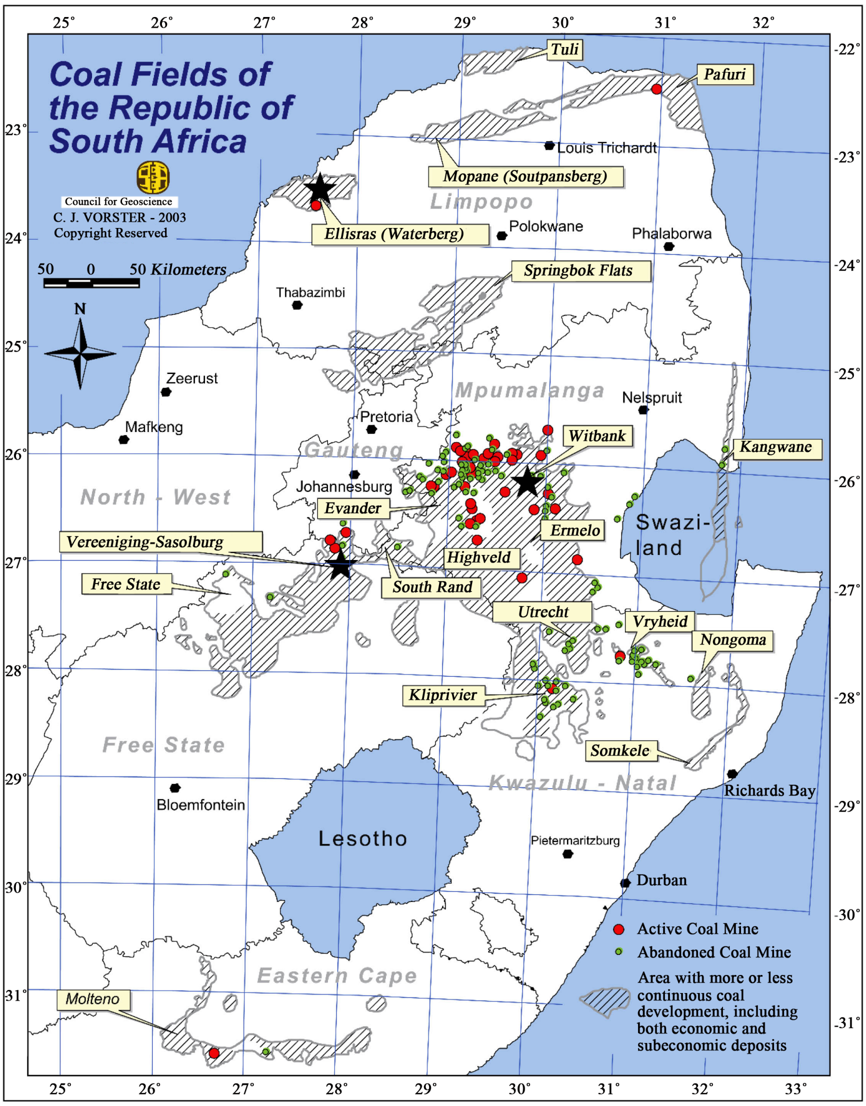

Figure 1. Coalfields of the Republic of South Africa [18]. The location of the CFPP are indicated.

ISO 13909:2001 [20]. Therefore, the quality of coal was concluded based on the ash yield and classified accordingly as indicated in **Table 1**.

In order to determine the radionuclides of interest (K^{40}, Ra^{226}, Th^{232} and Po^{210}) in the samples, two routes were followed. For K^{40}, Ra^{226} and Th^{232}, gamma spectrometry was conducted; for Po^{210} alpha spectrometry was conducted.

For gamma spectrometry, the coal and ash samples were dried for 24 hours in an air-circulation oven at 110˚C. Samples were further pulverized to obtain a fine powder and sieved for homogeneity. Thereafter, 100 g of each sample was placed in plastic containers of 6.5 cm diameter × 7.5 cm height, and sealed to make them airtight. The samples were left for a period of 1 month in a designated laboratory cupboard to ascertain the establishment of secular equilibrium between Ra^{226} and Th^{228} with their progeny and to prevent Rn loss. The specific radionuclides of the samples—*i.e.* K^{40}, Ra^{226} and Th^{232}—were determined using a

Table 1. Ash categorisation of the feed coals.

CFPP location	Ash Yield	Ash Class Category (According to ISO 11760:2005) [21]
Witbank	25.27% (≥20 and <30)	Moderately high ash
Waterberg	32.84% (≥30 and <50)	High ash
Sasolburg	40.85% (≥30 and <50)	High ash

high-resolution, p-type coaxial HPGe γ-ray spectrometer shielded by cylindrical lead. The detector relative efficiency was 28.2% and energy resolution of 1.67 keV-FWHM at the 1.33 MeV peak of Co^{60}. A cylindrical multi-nuclide source was used for detector energy calibration and efficiency determination [22]. The measured detection efficiencies were fitted by using a polynomial fitting function, as described by Khandaker *et al.* [23], and the fitted efficiencies were used in activity determination of the samples. The minimum detectable activity (MDA) of the γ-ray measurement system at 95% confidence level was calculated according to the procedure by Khandaker *et al.* [23]. Each sample was counted for 86,400 s, and similarly for background counts, in order to obtain the net activity.

The same method used by Sahu *et al.* [10] was used to determine the Po^{210} using alpha spectrometry. Samples (5 g of each) were first digested with HNO_3 and 4 N HCl sequentially. The acids were evaporated to near dryness and made up to 80 mL by adding 1 N HCl with ascorbic acid to reduce interfering Fe(III). Tracer activity 3.0 Bq/ml including Po^{209} was added to the aliquots. Then silver planchets were submerged into the solutions and were kept at a temperature of about 85˚C for 7 h with continuous stirring. The planchets thereafter were dried under infra-red lamp and alpha activities were determined in the alpha spectrometer. Samples were counted in an eight-chamber integrated alpha spectrometry system equipped with ion-implanted Si-charged-particle detectors, with an active detector-surface area of 450 mm^2, and a source-to-detector distance of approximately 10 mm. Samples and blanks were counted for nominally 250,000 s. Background measurements were made immediately prior to the measurements.

3. Results and Discussion

Table 2 presents the quality of coal used in the three CFPP and their coal and resultant coal combustion residues' radionuclide concentration in comparison with other studies [24] [25].

3.1. Radionuclide Concentrations in Coal, Fly Ash and Bottom Ash

The quality of coal used in South Africa varies between the CFPP [26], and is generally of low grade. The values obtained here are comparable to those reported by [27] for coals typically supplied to these three CFPP's. The quality of coal used can be classified as moderately high ash to high ash coal according to

Table 2. Ash categorisation of the feed coals and radionuclide concentrations in coal, fly ash and bottom ash for the three CFPP in comparison with other studies [24] [25].

Sample		Ash	N	Activity of radionuclide (Bq/kg)			
				K^{40}	Ra^{226}	Th^{232}	Po^{210}
Witbank (Present Study)	Fuel (Coal)	25%	9	88 ± 6	21 ± 1	22 ± 4	46 ± 1
	Fly ash		9	348 ± 8	80± 3	88 ± 6	193± 8
	Bottom ash		9	334 ± 8	63± 3	84 ± 6	41 ± 7
Waterberg (Present Study)	Fuel (Coal)	33%	9	91 ± 3	52 ± 5	21 ± 1	70 ± 5
	Fly ash		9	319 ± 4	166 ± 8	63 ± 6	280 ± 8
	Bottom ash		9	302 ± 5	150 ± 13	82 ± 8	3 ± 1
Sasolburg (Present Study)	Fuel (Coal)	41%	9	110 ± 4	24 ± 5	19 ± 3	83 ± 3
	Fly ash		9	286 ± 4	65± 6	45 ± 6	265 ± 13
	Bottom ash		9	242 ± 13	51± 6	44 ± 2	2 ± 1
Spain: Teruel UPT [25]	Fuel (Coal)	22%	2	66 ± 15	54 ± 3	21 ± 3	65 ± 11
	Fly ash		2	306 ± 13	191 ± 9	74 ± 3	257 ± 30
	Bottom ash		2	235 ± 11	149 ± 6	66 ± 3	57 ± 7
Spain: Litoral UPT [25]	Fuel (Coal)	16%	2	70 ± 16	15 ± 3	13 ± 3	33 ± 4
	Fly ash		2	338 ± 16	107± 4	87 ± 4	300 ± 40
	Bottom ash		2	278 ± 16	81 ± 4	64 ± 4	7 ± 1
Spain: Compostilla II [24]	Fuel (Coal)	37%	2	334 ± 13	34 ± 3	33 ± 3	118 ± 40
	Fly ash		2	1109 ± 49	94 ± 4	93 ± 4	416 ± 57
	Bottom ash		2	1077 ± 48	86 ± 2	89 ± 3	24 ± 12
Spain: Teruel [24]	Fuel (Coal)	28%	2	77 ± 32	56 ± 5	21 ± 2	65 ± 11
	Fly ash		2	310 ± 14	190 ± 2	74 ± 2	257 ±30
	Bottom ash		2	238 ± 13	149 ± 5	66 ± 3	57 ± 7
Spain: Litoral [24]	Fuel (Coal)	15%	4	62 ± 30	18 ± 4	20 ± 6	47 ± 20
	Fly ash		4	250 ± 144	158 ± 48	154 ± 53	505 ± 321
	Bottom ash		4	224 ± 105	144 ± 55	138 ± 61	9 ± 4

ISO [21]. For all of the CFPP samples, the radionuclide concentrations in the coals are in close proximity with the world averages (K^{40}, 4 - 785 Bq/kg; Ra^{226}, 1 - 206 Bq/kg ; Th^{232}, 1 - 170 Bq/kg; and Po^{210}, 3 - 52 Bq/kg) as indicated by the IAEA [28].

Although the radionuclide concentration in coal may be in line with the world averages, the concentrations of these are expected to be multiple times higher in the coal combustion products (fly ash and bottom ash) than the concentration in the original coal [12] and are commonly termed as the enrichment factor or enrichment ratio. The enrichment ratio is the concentration of the nuclide in ash to its concentration in the feed coal [10]. The results of radionuclides in the fly ash and bottom ash from **Table 2** certainly indicate the enrichment of radionuclides from the coal to the fly ash and bottom ash. The activity levels of K^{40}, Ra^{226}, and Th^{232} are very similar to each other in both the fly ash and bottom ash samples for all three plants in the current study. These correlate with the results from the two other studies tabulated. However, there are significant differences between the Po^{210} concentrations in the fly ash and bottom ash. This is because most of the Po^{210} is vaporized during combustion in the boiler [25]. This phenomenon will be expanded on in the next section. The K^{40}, Ra^{226}, Th^{232} and Po^{210}activity concentration levels for the fly ash and bottom ash for this study are in close proximity to the values reported by Mora *et al.* [25] and Baeza *et al.* [24], which are presented in **Table 2**. The average activity levels given in the UNSCEAR report [29] for fly ash are 265 Bq/kg for K^{40} , 240 Bq/kg for Ra^{226} , 70 Bq/kg for Th^{232} and 1700 Bq/kg for Po^{210}.

3.2. Enrichment Factors and Coal Quality

The enrichment factor for each radionuclide for the fly ash and bottom ash for each of the CFPP is reported in **Table 3** together with the results from other studies [24] [25].

Table 3. Coal quality and their respective enrichment factors for the three CFPP in comparison with other studies [24] [25].

Sample		Ash	Enrichment Factor			
			K^{40}	Ra^{226}	Th^{232}	Po^{210}
Witbank	Fly ash	25%	3.9 ± 0.8	3.8 ± 0.3	4 ± 0.7	4.1 ± 0.1
	Bottom ash		3.8 ± 0.8	3 ± 0.3	3.8 ± 0.7	0.9 ± 0.1
Waterberg	Fly ash	33%	3.5 ± 0.8	3.1 ± 0.6	3 ± 0.2	4 ± 0.6
	Bottom ash		3.3 ± 0.6	2.9 ± 0.4	3.9 ± 0.1	0
Sasolburg	Fly ash	41%	2.6 ± 1	2.7 ± 0.8	2.4 ± 0.5	3.2 ± 0.2
	Bottom ash		2.2 ± 0.3	2.1 ± 0.8	2.3 ± 0.7	0.0
Spain: Teruel UPT [25]	Fly ash	22%	4.6 ± 0.9	3.5 ± 0.3	3.5 ± 0.2	4 ± 0.3
	Bottom ash		3.6 ± 0.7	2.8 ± 0.7	3.1 ± 0.1	0.9 ± 0.6
Spain: Litoral UPT[25]	Fly ash	16%	4.8 ± 0.1	7.1 ± 0.8	6.7 ± 0.1	9 ± 0.1
	Bottom ash		4 ± 0.1	5.4 ± 0.8	4.9 ± 0.1	0.2 ± 0.3
Spain: Compostilla II [24]	Fly ash	37%	3.3 ± 0.2	2.8 ± 0.3	2.8 ± 0.3	4 ± 1
	Bottom ash		3.2 ± 0.2	2.5 ± 0.2	2.7 ± 0.3	0.2 ± 0.1
Spain: Teruel [24]	Fly ash	28%	4 ± 2	3.4 ± 0.3	3.5 ± 0.3	4 ± 1
	Bottom ash		3 ± 1	2.7 ± 0.3	3.1 ± 0.3	1 ± 0.2
Spain: Litoral [24]	Fly ash	15%	4 ± 3	9 ± 3	8 ± 3	11 ± 8
	Bottom ash		4 ± 2	8 ± 3	7 ± 4	0.2 ± 0.1

3.2.1. Fly Ash and Coal Quality

In most cases, the enrichment factors in this study were approximately 2 - 4 times higher in the ashes than in the feed coal. This is commonly observed in other studies [10] [30]. The enrichment factor for all the samples were higher for the fly ash compared to the bottom ash. This is because the radionuclides concentrate on the smaller fly ash particles that have a larger surface area-to-volume ratio and the hot flu gases cool down on their way to stack [31]. An exception to the enrichment of 2 - 4 times is that of Po^{210} in the bottom ash.

For the present study, the highest enrichment factor was for Po^{210} in the Witbank fly ash. The enrichment of Po^{210} in the fly ash is 2 - 4 times higher. The lowest enrichment factor in the fly ash was Th^{232} that was for the Sasolburg coal. In bottom ash the highest enrichment factor of 3.90 was observed for Th^{232} in the Waterberg coal and the lowest enrichment factor was 2.13 for Ra^{226} in the Sasolburg coal. In all cases, the fly ash had higher enrichment factors for all the radionuclides when compared to bottom ash, which is consistent with previous studies conducted [10] [32] [33].

The enrichment factor for each radionuclide determined in the fly ash (**Table 3**) was plotted against the ash content of the coal in order to establish a correlation (**Figures 2-6**).

It is apparent (from **Figures 2-5**) that the enrichment factor for K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash samples decreased as the %ash increased. This indicates that the enrichment factors for these specific radionuclides in these coals are directly proportional to coal quality as expressed by ash. A strong correlation (indicated by the R^2 values which are approximately > 0.7 in **Figures 2-5**) was obtained for the concentration the K^{40}, Ra^{226} and Th^{232} radionuclides in relation to the quality of coal, whilst the correlation for Po^{210} was moderately strong *i.e.* $R^2 = 0.62634$.

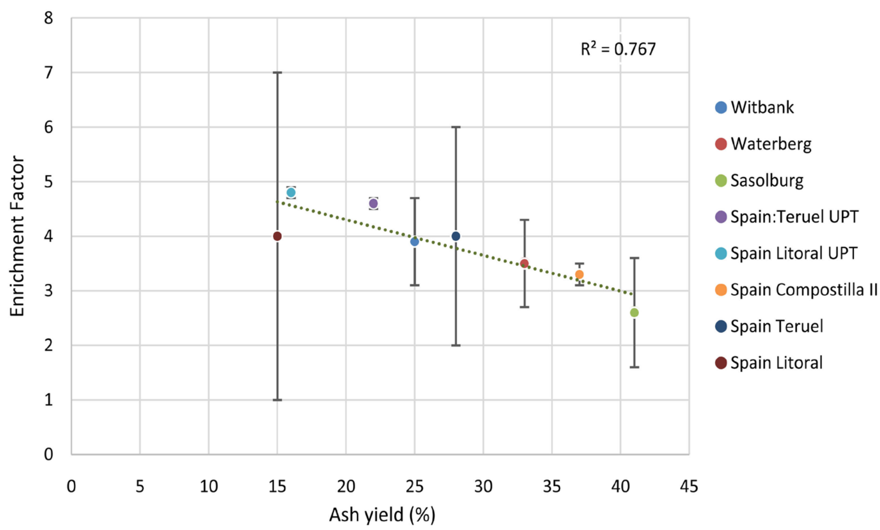

Figure 2. Correlation for K^{40} in the fly ash for the enrichment factor and the ash content in various coal samples.

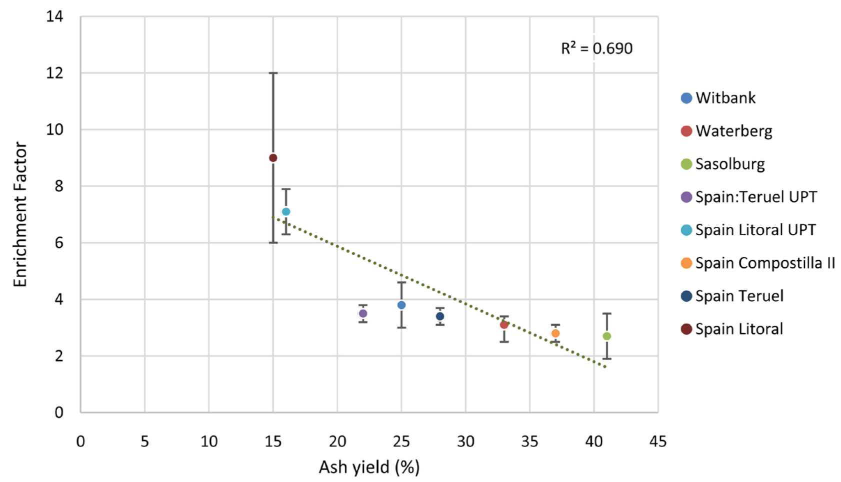

Figure 3. Correlation for Ra^{226} in the fly ash for the enrichment factor and the ash content in various coal samples.

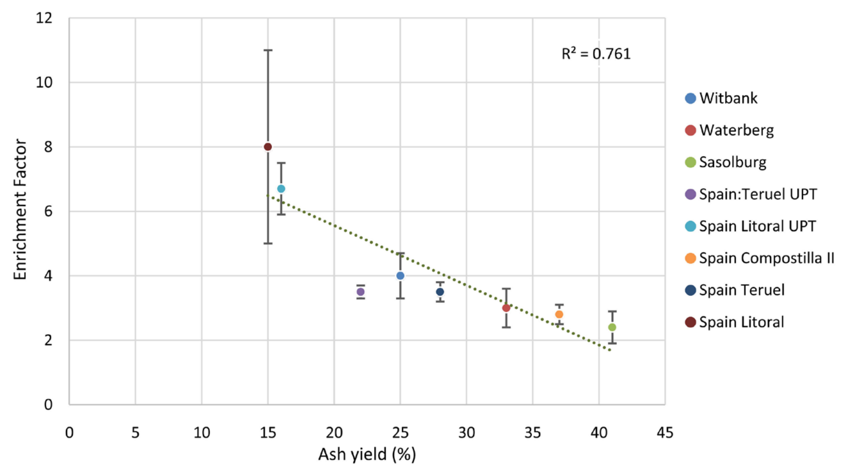

Figure 4. Correlation for Th^{232} in the fly ash for the enrichment factor and the ash content in various coal samples.

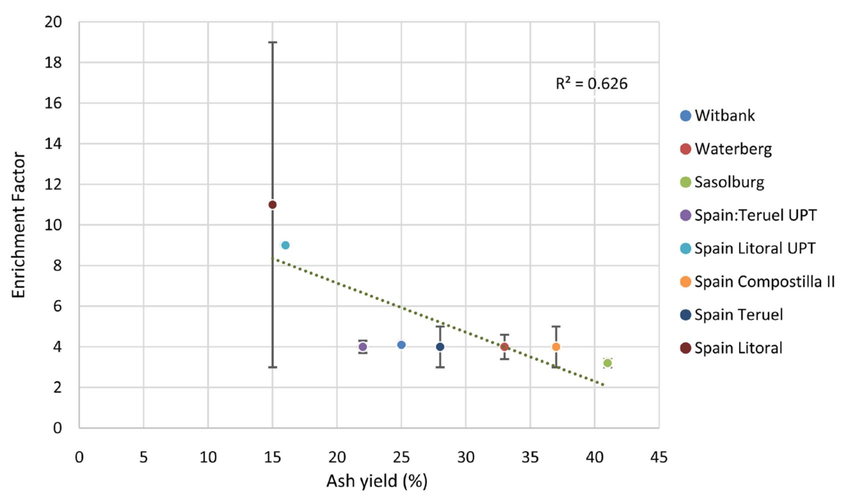

Figure 5. Correlation for Po^{210} in the fly ash for the enrichment factor and the ash content in various coal samples.

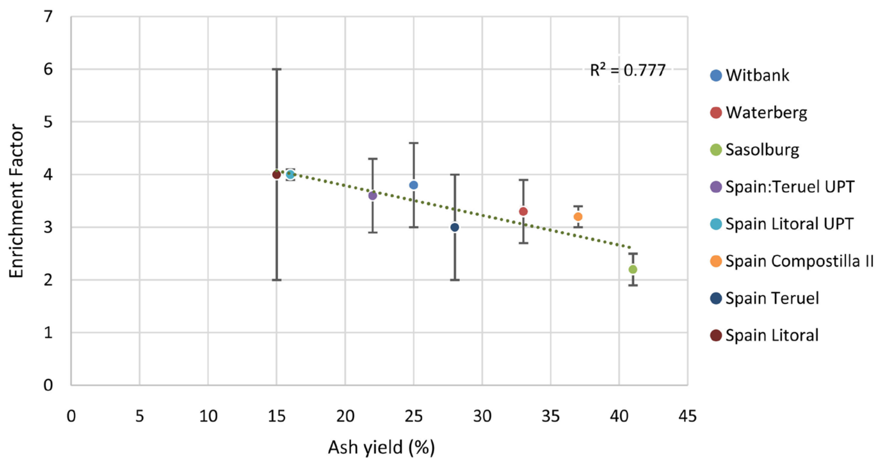

Figure 6. Correlation for K^{40} in the bottom ash for the enrichment factor and the ash content in various coal samples.

3.2.2. Bottom Ash and Coal Quality

The enrichment factors for each radionuclide in the bottom ash (Table 3) was correlated with the ash content of coal as shown in Figures 6-9.

In terms of the bottom ash, the enrichment factors also seems to be directly proportional to the ash content. However, a strong correlation is only present in K^{40} (R^2 = 0.7777) whereas the Ra^{226} and Th^{232} present only a moderately strong correlation (as opposed to strong correlations in the fly ash). It is interesting to note that this relationship is applicable to all the radionuclides (K^{40}, Ra^{226} and Th^{232}) except Po^{210}. In fact, Po^{210} exhibits no correlation (R^2 = 0.0854) and the enrichment factors for Po^{210} in the bottom ash for all the CFPP is less than unity, and therefore indicates that Po^{210} is depleted from the feed coal to bottom ash. This is because, as mentioned, the Po compounds are associated with sulphide minerals and are mostly volatilized during the combustion process; they later condense onto smaller fly ash particles which have larger specific areas and thus their levels get elevated in the fine fly ash fraction [10].

The results indicate that the enrichment factors for the radionuclides K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash and bottom ash are in close proximity with each other (except Po^{210} in the bottom ash). The results indicate that the enrichment factor is approximately 4, 3 and 2.4 when the ash content is 25%, 33% and 41% respectively. Mora *et al.* [25] observed that the enrichment factors in fly ash in relation to coal were 6.1 and 4.6 when the ash content in coals was 16% and 22%respectively. Lauer *et al.* [4] conclude that a 7 - 10 fold enrichment is expected from the elimination of carbon during combustion from coals containing 10% - 15% ash content, which is typical for low ash U.S coals. This leads to the following equation to determine the enrichment factor for radionuclides in coal prior to the combustion of the coal by simply considering the % ash content:

$$\text{Enrichment Factor} = \frac{1}{\text{Ash}(\%)\,\text{in feed coal}} \quad (1)$$

Although not precise, the equation may be used as an excellent estimate (or

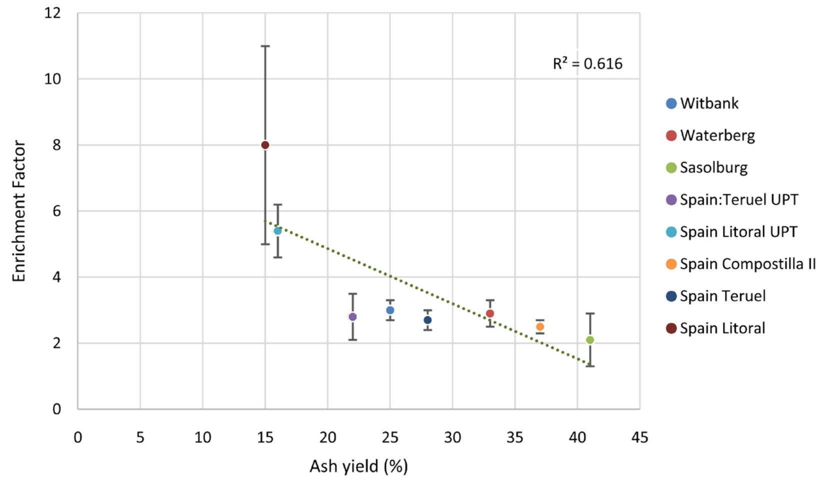

Figure 7. Correlation for Ra^{226} in the bottom ash for the enrichment factor and the ash content in various coal samples.

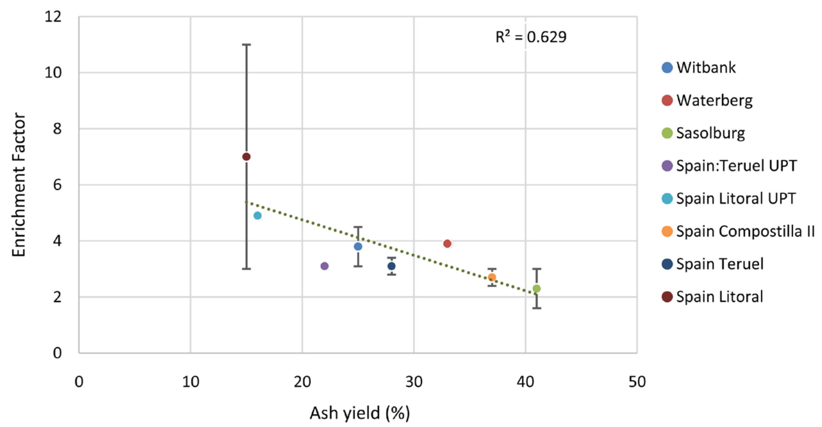

Figure 8. Correlation for Th^{232} in the bottom ash for the enrichment factor and the ash content in various coal samples.

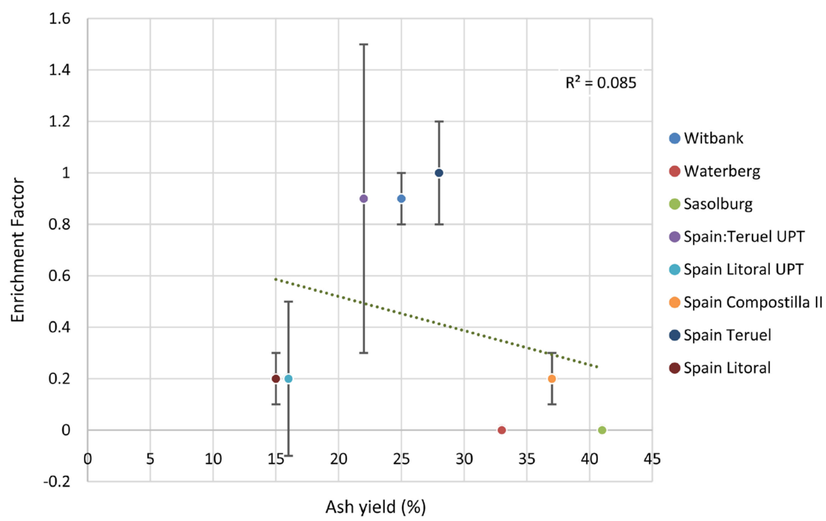

Figure 9. Correlation for Po^{210} in the bottom ash for the enrichment factor and the ash content in various coal samples.

rule of thumb) in determining the enrichment of radionuclides such as K^{40}, Ra^{226} and Th^{232} in coal combustion products.

4. Conclusions

When coal is burnt, the radionuclide concentration increases multiple times from the coal to the fly ash and bottom ash. The relationship between this enrichment of radionuclides (in the fly ash and bottom ash) and the quality of coal was investigated using samples from 3 different CFPP in South Africa in comparison with other studies.

It was found that the coals used in these CFPP were typically low quality coal, ranging between 25% - 45% ash content. The enrichment factor for the radionuclides K^{40}, Ra^{226}, Th^{232} and Po^{210} were 2 - 4 times higher in the ashes than in the feed coal (with the exception of Po^{210} in the bottom ash). The enrichment factor for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} and in the fly ash was found to be directly proportional to the ash content; *i.e.* when the ash % increased (coal quality decreases) the enrichment factor decreased. The relationship for the enrichment factor for the radionuclides K^{40}, Ra^{226} and Th^{232} in the bottom ash also showed the same directly proportional relationship to coal quality *i.e.* as the coal quality decreased the enrichment factor also decreased. However, this was not obeyed for the Po^{210} radionuclide, which had an enrichment factor less than unity in the bottom ash samples.

The relationship between coal quality and enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232}, and Po^{210} in both the fly ash and bottom ash (except Po^{210} in the bottom ash) was demonstrated by using the following mathematical equation:

$$\text{Enrichment Factor} = \frac{1}{\text{Ash}\left(\%\right)\text{in feed coal}} \tag{1}$$

This equation may be used a good indication in obtaining a ball park figure in determining the enrichment of radionuclides such as K^{40}, Ra^{226} and Th^{232} in coal combustion products such as fly ash and bottom ash.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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