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Natural Radioactivity Releases from Lignite Power Plants in Greece

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ABSTRACT

In Greece, power stations burning lignite from two large deposits, in the regions of Megalopolis and Ptolemais, play an important role in meeting the electric energy needs of the country. The investigation reported here deals with the determination of the ^{226}Ra, ^{228}Ra and ^{40}K concentrations in the lignite feeding two power units in Megalopolis and in the product ash. Systematic sampling allowed evaluation of mean representative concentrations and estimation of the resulting atmospheric discharges of radioactivity. Furthermore, statistical analysis has shown that, at the 95% confidence level, essentially all ^{226}Ra from the lignite feeding each unit is accounted for in the ash. The results of both the present and previous investigations on two units burning lignite from the Ptolemais deposit show that electricity generation from Greek lignites is associated with radioactivity discharges which are rather high relative to corresponding discharge data from the literature.

INTRODUCTION

Lignite plays an important role within the electric power system of Greece. In 1985, the lignite power stations contributed almost 65% of the total electric energy generated. There are two very important lignite deposits under exploitation. The first, in the northern part of the country, is the Ptolemais deposit which, from various fields, supplies three local power stations with a total installed capacity of 3020 MW. The second is located in the centre of the Peloponese peninsula near the village of Megalopolis and supplies a three-unit power station (2×125 MW and 1×300 MW). A

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fourth 300-MW unit is now under construction. The Megalopolis lignite is of very low calorific value and has rather high water and ash contents—it is amongst the poorest burned for generation of electricity. The local power station consumes approximately 2·3 kg of natural lignite per kWh produced.

The purpose of the investigation reported here was to determine representative values for the ^{226}Ra, ^{228}Ra and ^{40}K concentrations in the lignite burned and the ash produced by two units (125 and 300 MW) of the Megalopolis power plant, which is fed with surface-mined lignite from the nearby deposit. Since the concentrations of these radionuclides may vary, even quite abruptly within a single lignite seam (Corbett, 1983), this determination required extensive sampling and analysis. A similar investigation of the Ptolemais deposit has already been performed by the authors (Simopoulos & Angelopoulos, 1986).

The concentrations of ^{226}Ra, ^{228}Ra and ^{40}K can easily be determined by γ-spectroscopy. The results can be used to assess the source term for estimation of the radiological impact of a coal-fired power station. They are also of interest in the context of product ash use in building materials.

SAMPLING AND PREPARATION OF SAMPLES

At each unit investigated, samples of (a) the lignite feeding the boiler, (b) fly ash from the electrostatic precipitator (ESP) and (c) bottom ash, were collected daily and for five consecutive weeks. The water and ash contents, as well as the calorific value of each lignite sample, were determined, while for each unit records were maintained of the daily consumption of natural lignite and of the electric energy produced. During the sampling period, the output of the units was kept as constant as possible, in order to limit any effect on the partitioning of the measured radionuclides.

The samples collected were used to prepare five 'weekly specimens' for each of the materials examined. A lignite specimen was formed by mixing quantities of the daily samples, proportional by weight to the quantity of natural lignite consumed each day by the respective unit. Such a specimen can be considered representative of the average natural lignite fed to the unit during the relevant week. A fly or bottom-ash specimen was similarly produced by mixing quantities of the daily samples, proportional by weight to the ash content and to the quantity of natural lignite consumed each day by the respective unit. Furthermore, all specimens were dried under ambient temperature and the lignite pulverized to less than 90 μm. Finally, the water contents of all specimens were determined. This preparatory work was performed by personnel of the Megalopolis Power Plant (Drosatos, 1984). Table 1 presents the characteristic data for the lignite burned by each of the units during the five-week sampling period.

TABLE 1
Characteristic Data for the Lignite Burned during the Sampling Period (number of weekly specimens = 5)

Natural lignite	*Unit I (125 MW)*	*Unit III (300 MW)*
Water content (%)		
max	56·8	59·4
min	56·2	56·4
mean	56·6	57·4
Ash content (%)		
max	19·1	19·0
min	18·2	16·1
mean	18·5	17·8
Specific consumption (kg/kWh)	2·32	2·31

Each weekly specimen was used to fill two plastic cylindrical boxes, 75 mm in diameter and 70 mm high. The boxes were sealed and covered with a film of epoxy resin to limit, as far as possible, escape of radon. Thus, in each case, duplicate specimens were analysed.

EXPERIMENTAL PROCEDURE

The concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in the samples were determined by γ-spectroscopy. Two systems were used, each consisting of a shielded detector connected to a 4096 multichannel analyzer on-line to a PDP-11/04 computer operating under RT11-CLASS/Spectran-III software (Canberra, 1978). The detectors were a coaxial Ge(Li) (eff. 23·8%, resoln. 1·97 keV FWHM at 1·33 MeV, Peak/Compton 47:1 at 1·33 MeV) and a coaxial pure Ge (eff. 33·8%, resoln. 1·78 keV FWHM at 1·33 MeV, Peak/Compton 66·5:1 at 1·33 MeV). The energy and efficiency calibrations of the detector systems were performed with a certified mixed radionuclide solution providing eleven principal γ-rays in the range 90 to 1800 keV. Since the collected spectra were complex, the multiplet analysis method was applied for analysis. Thus, when the analysis detected multiplets, a non-linear least-squares fit of Gaussian functions, one for each suspected component, was invoked to determine the contribution to the background-subtracted multiplet area. The uncertainty in the area of each peak, at 90% confidence level, was determined by counting statistics in the case of a singlet or by both counting statistics and the degree of data fit by the assumed Gaussian

components in the case of a multiplet. All data required for the analysis of the photopeaks were taken from Erdtman & Soyka (1979).

To allow for equilibrium of ^{226}Ra and ^{228}Ra with their decay products, all specimens were analyzed at least three weeks after the boxes were sealed. Of course, it can be argued that, despite thorough sealing of the boxes, some radon could still escape; however, the observation that boxes, sealed four years ago, showed a marked deformation owing to the pressure developed by the product gases justifies the assumption of a satisfactory equilibrium. The counting time was not the same for all measurements. Since ^{226}Ra was considered the most important radionuclide in the samples, it was essential to limit the uncertainty in the estimation of its concentration from the less-efficient photopeak of the daughter radionuclides in equilibrium, from the points of view both of detector efficiency and isotope yield. Thus the ca. 1·76 MeV photopeak of ^{214}Bi was selected for this purpose and counting was ended automatically when the uncertainty was less than 3%, leading to counting times in the range 5 to 20×10^4s. With this uncertainty, the total error associated with the measurement of ^{226}Ra is estimated to be less than 6%. The errors on ^{228}Ra and ^{40}K assays were larger because, in all samples, these nuclides were present at much lower concentrations than ^{226}Ra. Finally, no self-absorption correction was applied on the assumption that the standard solution used for calibration and the specimens examined produce, under identical geometries, similar photopeak attenuation. The ^{226}Ra concentrations have been derived from the weighted mean of the activities of two photopeaks of ^{214}Pb (295·2, 352·0 keV) and of three photopeaks of ^{214}Bi (609·3, 1120·9, 1764·5 keV). Direct determination of ^{226}Ra concentrations from the 186 keV photopeak, after appropriate correction for the contribution of the 185 keV photopeak of ^{235}U, has not been attempted, mainly because of the large range of literature values for the yield of ^{226}Ra at this energy (Chouak *et al.*, 1978 and Erdtman & Soyka, 1979).

For ^{228}Ra, two photopeaks of ^{228}Ac (338·7, 911·1 keV) and the photopeaks of ^{212}Pb (238·6 keV) and ^{208}Tl (583·1 keV) were used in the same way. Finally, the concentration of ^{40}K was obtained from the single photopeak of this isotope at 1460·8 keV.

RESULTS

Table 2 summarizes the mean values and the standard deviations of the ^{226}Ra, ^{228}Ra and ^{40}K concentrations in lignite, fly ash (ESP) and bottom ash for each unit. The results are based on the respective concentrations in the five weekly specimens, evaluated as previously explained.

TABLE 2
Mean Concentrations (Bq/kg) of Dry Material ($\pm 1\sigma$)

Power unit	*No. samples/ specimens*	*Deposit*									*Reference*
		Lignite			*Fly ash*			*Bottom ash*			
		^{226}Ra (^{238}U)	^{228}Ra (^{232}Th)	^{40}K	^{226}Ra (^{238}U)	^{228}Ra (^{232}Th)	^{40}K	^{226}Ra (^{238}U)	^{228}Ra (^{232}Th)	^{40}K	
Megalopolis											
Unit I	35/5	314 ± 52	21 ± 2	181 ± 26	807 ± 138	55 ± 2	449 ± 47	546 ± 77	44 ± 3	406 ± 63	This work
Unit III	35/5	321 ± 40	21 ± 1	191 ± 14	845 ± 71	56 ± 1	502 ± 21	587 ± 71	44 ± 2	423 ± 30	
Ptolemais											
Ptolemais IV	31/5	83 ± 6	15 ± 1	78 ± 13	261 ± 19	45 ± 2	251 ± 16	114 ± 16	24 ± 3	137 ± 21	Simopoulos &
Kardia I	33/5	175 ± 20	16 ± 2	68 ± 13	600 ± 75	49 ± 5	217 ± 17	363 ± 29	36 ± 2	200 ± 11	Angelopoulos, 1986
USA											
S. Dakota	–	9	4	–	161	84	–			–	Styron *et al.*,
Minnesota	–	16	7	–	137	67	–	-	–	–	1981
France											
Power station (mean)	–	40	(5)	17	–	–	–		–	–	Aigueperse
55 MW unit	–	–	–	–	370[a]	(40)[a]	220[a]	–	–	–	*et al.*, 1982
250 MW unit	–	–	–	–	520[a]	–	–	–	–	–	
FRG											
600 MW unit	–	–			63	22	333		–	–	Chatterjee *et al.*, 1979
Italy											
Central (range)	–	4–15	(74–111)	–	37–74	(333)[b]	–	-	–	–	Mastinu, 1979
Sardinia	–	(252)	–	–	(999)	–	–	-	–	–	

[a] Fly ash collected in the stack.
[b] Fly ash from the last stage of the ESP.

It should be pointed out that the units investigated are fed with a mixture of lignite originating from two different fields of the Megalopolis deposit. The quantities mixed are variable and different for each unit. Furthermore, depending on the quality of the lignite, the firing of the boilers is occasionally supported by negligible quantities of oil. Samples of the oil used during the sampling period have been examined but they did not show any detectable radioactivity.

Measurements of the radioactivity of Greek lignites and of ash samples from various power stations in Greece have been reported by Papastefanou & Charalambous (1979, 1980, 1983). The values for Megalopolis, as they appear in these publications, are:

Lignite
^{226}Ra, $3{\cdot}2 \pm 0{\cdot}7$ pCi g^{-1} (118 ± 26 Bq kg^{-1})
^{228}Ra, $0{\cdot}023 \pm 0{\cdot}005$ pCi g^{-1} ($0{\cdot}85 \pm 0{\cdot}19$ Bq kg^{-1})
Fly ash
^{226}Ra, $10{\cdot}6 \pm 0{\cdot}9$ pCi g^{-1} (392 ± 33 Bq kg^{-1})
^{228}Ra, $0{\cdot}193 \pm 0{\cdot}026$ pCi g^{-1} (7 ± 1 Bq kg^{-1})
Bottom ash
^{226}Ra, $8{\cdot}2 \pm 0{\cdot}9$ pCi g^{-1} (303 ± 33 Bq kg^{-1})

The above concentrations are much lower than those obtained in the present investigation. However, this comparison cannot be interpreted further because no details are given by Papastefanou & Charalambous on sampling procedure, extent of sampling and measures taken to ensure that the ash samples examined corresponded to the ash produced by the power station when burning the sampled lignite.

Table 2 also includes the results obtained by the present authors here (Simopoulos & Angelopoulos, 1986) during a previous similar investigation of two units supplied with lignite from the Ptolemais deposit, which is the largest in use in Greece for electricity generation. In this case, the lignite burned by each unit (Ptolemais–IV and Kardia–I) originated from a different field of the deposit. For comparison, Table 2 also presents some published data on radioisotope concentrations in lignite and fly ash as measured in other countries (Chatterjee *et al.*, 1979; Mastinu, 1979; Styron *et al.*, 1981; Aigueperse *et al.*, 1982).

ENRICHMENT RATIOS AND PARTITIONING COEFFICIENTS OF ^{226}Ra

The systematic nature of sampling and the measures introduced to limit the statistical error in ^{226}Ra estimation allow comparison of the concentrations

of this radionuclide in lignite and ash in terms of the enrichment ratio as defined by Styron *et al.* (1981). According to this definition, the enrichment ratio of an isotope is defined as the ratio of its concentration in a given ash fraction to its ash equivalent concentration in the lignite. Thus, the ^{226}Ra enrichment ratio (ER_i) for fly ash (ER_f) and for bottom ash (ER_b) are expressed as follows:

$$ER_i = \frac{C_i}{C_l^*} \tag{1}$$

where i = f or b for fly or bottom ash respectively, C_i is the ^{226}Ra concentration in the ash and C_l^* is the ash equivalent concentration of ^{226}Ra in dry lignite. If C_l is the ^{226}Ra concentration of dry lignite and ω and τ the water and ash content of the natural lignite then $C_l^* = (1-\omega)C_l/\tau$ and hence:

$$ER_i = \frac{\tau C_i}{(1-\omega)C_l} \tag{2}$$

After lignite is burned, the contained radionuclides are partitioned between the resulting effluents. Following Styron *et al.* (1981), the radionuclide partitioning coefficient represents the percentage of the total amount of the radionuclide present in the lignite which is found in a particular effluent. Thus, if x denotes the fly-ash partitioning coefficient of the ash, i.e. the fly ash expressed as a weight fraction of the total ash, then, in the case of ^{226}Ra, the partitioning coefficients for fly ash (RPC_f) and for bottom ash (RPC_b) are:

$$RPC_f = x \cdot ER_f \text{ and } RPC_b = (1-x) \cdot ER_b \tag{3}$$

Finally, the ^{226}Ra total partitioning coefficient, $RTPC$, is defined as:

$$RTPC = RPC_f + RPC_b \tag{4}$$

Clearly, if the total amount of ^{226}Ra contained in the burned lignite is carried out by the product fly and bottom ash, then $RTPC$ should equal unity.

If R_l and R_{ash} are respectively the ^{226}Ra activities in 1 kg of natural lignite and in the ash resulting from its burning, i.e.

$$R_l = (1-\omega)C_l \text{ and } R_{ash} = x\tau C_f + (1-x)\tau C_b \tag{5}$$

then it is easy to verify that:

$$RTPC = \frac{R_{ash}}{R_l} \tag{6}$$

Equation (6) indicates that, in order to examine whether the total amount of ^{226}Ra contained in the burned lignite is discharged by the fly ash and bottom ash produced, instead of comparing *RTPC* to unity, one could compare R_l and R_{ash}. This radioactivity balance has been examined by application of a significance test, at the 95% confidence level, to the difference $R_l - R_{ash}$. For this test, R_l and R_{ash} have been calculated using the mean values and the standard deviations of the ^{226}Ra concentrations C_l, C_f and C_b obtained as being representative of the whole sampling period (Table 2). The results are shown in Table 3.

TABLE 3
Enrichment Ratios, Partitioning Coefficients and Radioactivity Balance for ^{226}Ra ($\pm 1\sigma$)

Unit investigated	*Unit I (125 MW)*	*Unit III (300 MW)*
Enrichment ratios		
fly ash (ER_f)	1·1 ± 0·26	1·1 ± 0·15
bottom ash (ER_b)	0·7 ± 0·16	0·8 ± 0·13
Partitioning coefficients		
fly ash (RPC_f)	0·9 ± 0·22	0·9 ± 0·13
bottom ash (RPC_b)	0·1 ± 0·02	0·1 ± 0·02
total (*RTPC*)	1·0 ± 0·22	1·0 ± 0·13
Radioactivity balance[a]		
lignite input radioactivity (R_l)	136 ± 23	137 ± 15
ash output radioactivity (R_{ash})	142 ± 22	144 ± 11
significance[b]	39	19

[a] In Bq per kg of natural lignite.
[b] Statistically-significant different ($R_{ash} - R_l$) at the 95% confidence level.

The data in Table 3 indicate that, for both units, the bottom ash is depleted in ^{226}Ra, whilst the fly ash is slightly enriched in this isotope. Furthermore, the results of the statistical test show that, at the 95% confidence level, the difference between the calculated values of R_l and R_{ash} is much lower than the statistically-significant difference $R_l - R_{ash}$. This finding for both units confirms the view expressed elsewhere (Styron *et al.*, 1981) that essentially all ^{226}Ra from feed lignite is accounted for in fly and bottom ash.

ATMOSPHERIC DISCHARGES AND THEIR RADIOLOGICAL SIGNIFICANCE

The radionuclide concentration data from this study can be used to estimate the radioactivity released by fly-ash escape from the stack of each unit per unit production (1 GWa) of electric energy. At both units, the fly ash produced was 85% of the total ash, while the efficiency of the ESP was 98% (Drosatos, 1984). Assuming now that the escaping fly ash contains the mean measured concentrations shown in Table 2 and using the consumption data of Table 1, the atmospheric discharges presented in Table 4 are derived.

TABLE 4
Atmospheric Discharges (MBq/GWa)

Nuclide	*Megalopolis*		*Ptolemais*		*Worldwide*[a]
	Unit I	*Unit III*	*Ptolemais-IV*	*Kardia-I*	
^{226}Ra	51 600	51 700	12 300	19 800	70–18 000
^{228}Ra	3 500	3 400	2 100	1 600	40–15 000
^{40}K	28 700	25 900	11 800	7200	670–20 000

[a] UNSCEAR, 1982.

The calculated discharges are tabulated along with those from the two units in Ptolemais previously investigated by the authors (Simopoulos & Angelopoulos, 1986) and with the range of estimated discharges published by UNSCEAR (1982). The comparison shows that atmospheric discharges from the four Greek lignite units investigated, especially from those in Megalopolis, are high. They can be reduced by improving the efficiency of the electrostatic precipitators.

The radiological significance of the results presented in this paper can be assessed by estimating the collective effective dose equivalent commitments arising from the discharges of the lignite power units. To a first approximation, this can be achieved by simple proportionation, using the dose data published by UNSCEAR (1982) for world-wide coal burning. These data have been obtained using, amongst others, the following assumptions which are relevant to the determination of source terms in the present case:

(i) In coal, all decay products of ^{238}U and ^{232}Th are in radioactive equilibrium with their precursors.
(ii) The fly-ash enrichment factors are 1 for the isotopes of radium, uranium and thorium, and 3 for ^{210}Pb and ^{210}Po.
(iii) ^{222}Rn and ^{220}Rn, being in radioactive equilibrium with ^{238}U and ^{232}Th respectively, are discharged in their entirety when coal is burned.

With these assumptions and using the arithmetic mean values of the measured concentrations (Table 2) and of the discharges presented in Table 4, the following source terms are derived for estimation, by simple proportionation, of the population doses arising from the combustion of the Greek lignites investigated (discharges in GBq $(GWa)^{-1}$):

^{238}U decay series, from ^{238}U to ^{226}Ra—33·9
^{210}Pb and ^{210}Po—100
^{232}Th decay series, from ^{232}Th to ^{224}Ra—2·7
^{222}Rn—1955
^{220}Rn—158

The radon discharges have been estimated taking into account that production of 1 kWh requires consumption of 2·3 kg of natural lignite as mentioned in the Introduction, i.e. roughly 1 kg of dry lignite with activity concentrations of ^{222}Rn and ^{230}Rn equal respectively 223 and 18 Bq kg^{-1}.

The above discharges from lignite burning, with respective reference to those values applied in UNSCEAR (1982), can readily be used to estimate population doses incurred via inhalation, ingestion and irradiation. The results, presented in Table 5, show that the total collective effective dose equivalent commitment from Greek lignites (23 man Sv/GWa) is much larger than the value, 2 man Sv/GWa, given by UNSCEAR (1982) for world-wide coal burning. The high value, however, is only 0·6% of the figure given by Corbett (1983) as the average natural dose to a population which consumes electrical power of 1 GW average.

TABLE 5
Collective Effective Dose Equivalent Commitments (10^{-2} man Sv/GWa)

Exposure route	*Greek lignites (this work)*	*Coal, worldwide*[a]
Inhalation during cloud passage		
uranium series	734·3	33
thorium series	187·6	104·2
Internal irradiation		
uranium series	1 244·6	45
thorium series	28·3	11·2
External irradiation due to the activity deposited		
uranium series	90	4
thorium series	9	5
Total	2 300	200

[a] UNSCEAR, 1982.

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