# On the Radioactivity of Fly Ashes from Coal Power Plants

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The radioactivity of Greek lignites and their ashes was studied. It was found that the radionuclides in them belong to the uranium series. Fly ashes are relatively rich in radioactivity. Concentrations up to $7 \cdot 10^{-5}$ g/g and 12 pCi/g were measured for $^{238}$U and $^{226}$Ra, respectively. The associated hazards from radioactivity escaping from stacks of thermopower plants or when fly ashes are used as subtitutes of cement in concrete are evaluated.

## 1. Introduction

As pointed out by Jaworowski et al. [1], the $^{226}$Ra concentration in atmospheric precipitates has increased by a factor of hundred during the last 80 years. The principal source of this increase beyond the natural Ra-226 concentration is coal burning. Coal contains Ra-226 and other radionuclides [2–4]. The greatest part of coal radioactivity remains with the ashes. Ra from coal is mostly introduced into the atmosphere as a constituent of fly ash. Fly ashes from thermopower plants escape into the atmosphere in quantities of about 0.5% to 2% of the total ash produced and constitute a potential hazard in the vicinity of the plant.

In many papers (see for example Venuat [5] and Voyatzakis et al. [6] it has been proposed to use fly ash in the production of concrete. This again would involve potential hazards arising either from direct irradiation from concrete or from the radon escaping from it.

In this paper the radioactivity of Greek lignites and their ashes is studied. The amount of Ra escaping from the stacks of plants is deduced and its hazard is estimated. Also, same estimations of the hazards were made should the ashes be used in cement.

## 2. Experimental Procedure and Results

The samples studied were from the principal Greek coal mines, i. e. from Ptolemais (Northern Greece), from Aliverion (Island Evia) and from Megalopolis (Peloponessus), where there are 2 (Kardia and LIPTOL), 1 and 1 thermopower plants, respectively. Each of these plants burns about $20 \cdot 10^3$ tons of lignite daily. These lignites are poor in coal. The average composition of Ptolemais lignites and of their ashes is given in the appendix. The lignites (directly from the mines and after drying) and the ashes after burning, i. e. "liquid" ashes and "fly" ashes, were measured for radioactivity.

The lignite samples were powdered in a mortar of checked low radioactivity. 40 g of these and the powders of liquid and fly ash were placed in small plastic containers (boxes) of 6 cm diameter of low radioactivity. The plastic boxes with the fly ashes were sealed in order to prevent Rn escape.

The radioactivity of the samples was mainly measured by a Ge-Li detector. The detector had a volume of 40 cm$^3$, resolution of about 2 keV at 1.33 MeV of $^{60}$Co, peak-to-Compton 25/1. Special studies were made to find the overall efficiency of the detector due to the particular size of the samples. The efficiency was known to an accuracy of better than 10%. Some samples were also measured by an intrinsic Ge detector. This detector had an active area of 200 mm$^2$, depletion depth of 5 mm and resolution of about 420 eV at 60 keV ($^{241}$Am). This detector was used for the investigation of the 63 keV ($^{238}$U) line.

Each sample was measured several times in intervals of about a week to observe the build-up of $^{214}$Bi. The duration of each measurement was $50 \cdot 10^3$ sec. Background measurements of the same duration were taken between the measurements of the samples.

In Fig. 1 a typical gamma spectrum for a sample of fly ash is presented. The background is not subtracted. In the upper right corner of the figure the spectrum of the background in the region 600 keV is given for comparison.

In general, we found in the coals and the ashes radionuclides of the uranium series, well above

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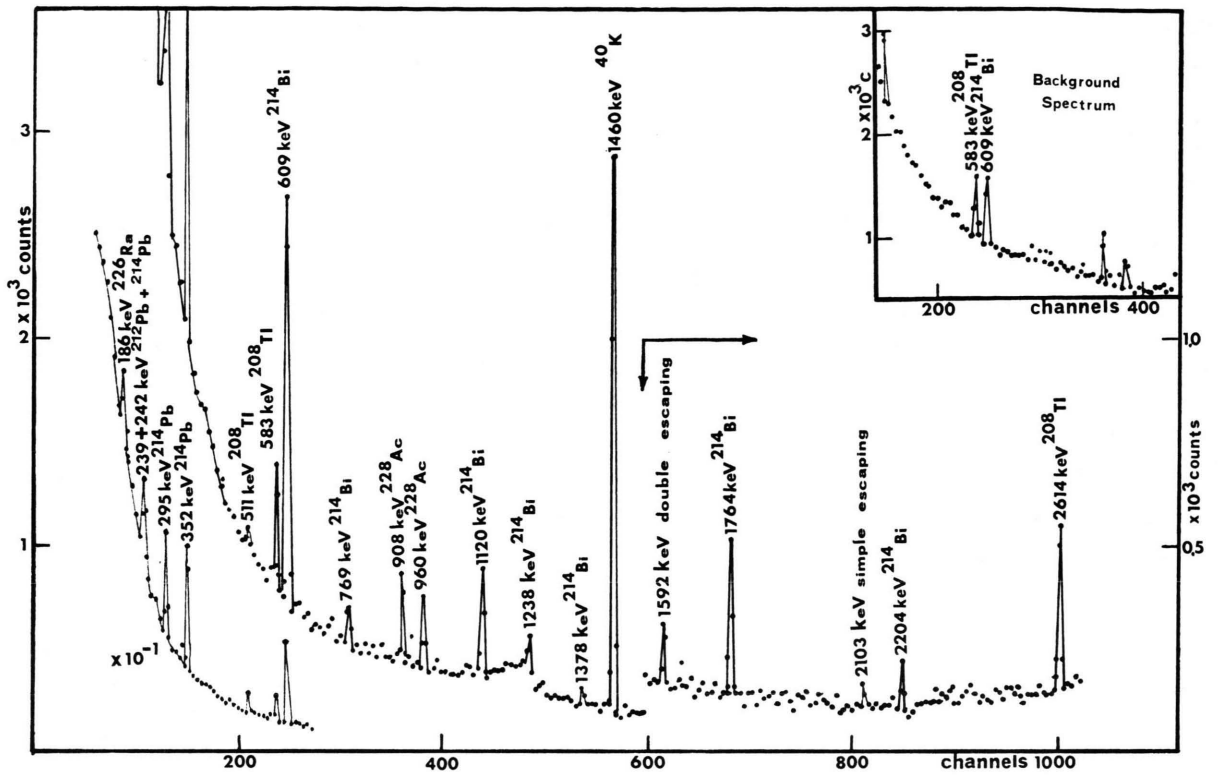


Fig. 1. Typical gamma spectrum of fly ash from Kardia, Ptolemais. The background is not subtracted.

background, while radionuclides of the thorium series were covered by the background, if they exist. $^{40}K$ is also covered by the background.

The specific activity of each radionuclide present in the samples measured is evaluated from the intensity of the lines after background subtraction, after taking into account the appropriate factors of detection efficiency of the system, the branching ratio of the transitions, etc. The results for $^{238}U$ and $^{226}Ra$ are presented in Table 1.

Table 1. Radioactivity of Greek Lignites and their ashes.

| Lab. No. | Kind of Sample | Region of Sampling | $^{238}U$ x $10^{-5}$ g/g sample | $^{226}Ra$ pCi/g sample | |
|---|---|---|---|---|---|
| | | | from 63 keV | from 186 keV | from 609 keV |
| L-3 | brown coal | Power Station Kardia | 3.4±1.0 | 6.9±0.8 | (0.8±0.7) |
| L-5 | ash (liquid) | Power Station Kardia | 3.9±1.0 | 4.7±0.8 | (4.6±0.7) |
| L-6 | fly ash | Power Station Kardia | 7.0±1.0 | 10.4±0.9 | 11.4±0.8 |
| L-7 | coke | Power Station Ptolemais | 2.6±1.0 | 3.9±0.7 | (0.2±0.7) |
| L-10 | fly ash | Power Station Ptolemais | 4.1±1.0 | 11.4±0.8 | 12.7±0.8 |
| L-11 | ash (liquid) | Nitrogenous Fertilizer Factory Ptolemais | 4.4±1.0 | 5.9±0.8 | (2.4±0.7) |
| L-12 | fly ash | Nitrogenous Fertilizer Factory Ptolemais | 3.7±1.0 | 5.7±0.7 | 4.3±0.7 |
| L-17 | brown coal | Power Station Aliveri | 2.4±1.0 | 3.6±0.7 | (1.4±0.7) |
| L-19 | ash (liquid) | Power Station Aliveri | 3.9±1.0 | 6.2±0.8 | (3.4±0.7) |
| L-20 | fly ash | Power Station Aliveri | 4.3±1.0 | 8.3±0.9 | 7.0±0.8 |
| L-23 | brown coal | Power Station Megalopolis | 3.5±1.0 | 3.2±0.7 | (2.3±0.7) |
| L-25 | ash (liquid) | Power Station Megalopolis | 5.9±1.0 | 8.2±0.9 | (5.2±0.8) |
| L-26 | fly ash | Power Station Megalopolis | 4.0±1.0 | 10.6±0.9 | 12.1±0.8 |

The specific concentration of $^{238}U$ in the samples is directly deduced from the activity of its first daughter $^{234}Th$ (63 keV line), while for $^{226}Ra$ the activity of the "186" keV line and the activity of the 609 keV line (transition of $^{214}Bi$) were used. For the latter radionuclide radioactive equilibrium between $^{214}Bi$ and $^{226}Ra$ was assumed. In the activity of the "186" keV line we have a contribution of about 10% from the 185 keV line of $^{235}U$, which was taken into account. Because of Rn escape the 186 keV line is the sure basis for the concentration of $^{226}Ra$. This line was used in our paper. The differences of $^{226}Ra$ concentration shown in columns 5) and 6) of Table 1 for the unsealed samples (that are in parentheses) are due to Rn escape. In the cases of sealed samples (fly ashes) a fair agreement between the calculations by 186 keV and through the 609 keV lines exists.

## 3. Remarks and Discussion

We found that Greek lignites contain a relatively large amount of U and its daughters and no other significant radionuclides. The measurements show that $^{226}Ra$ and $^{238}U$ in the lignites studied are not in equilibrium. This is probably due to a leaching of $^{226}Ra$. Leaching of $^{226}Ra$ from porous materials, for example from fossil bones, should be a general phenomenon [7].

The concentrations of both $^{238}U$ and $^{226}Ra$ in lignites and in their ashes were found to be in general higher than the published (to our knowledge) values [1, 4]. Equilibrium between $^{238}U$ and its daughters is not to be expected in the ashes as their relative concentrations will be a function of the burning temperature and the volatilization point of the different elements.

*a) Hazards from radioactivity escaping from stacks*

Fly ashes escape from the stacks of thermopower plants in large quantities (40 tons per day from each stack). The collection efficiency of the filters used varies. For the power plants studied, the escaping dust amounts to about 2% of the total ash produced. Assuming that the escaping dust has the same radioactive concentration as the measured fly ash and taking into account that $20 \cdot 10^3$ tons of lignite is burned per day and using the experimental data, we found that from the stack of each power plant escaped about $Q_0 = 500\ \mu Ci$ $^{226}Ra$ per day.

Radioactive effluents emitted from the stack of a thermopower plant behave in essentially the same way as ordinary smoke. The problem of ordinary smoke dispersion in the atmosphere is well studied in a monography by Pasquill [8] using the general diffusion theory. For our calculation we follow an analysis similar to that which was given by Lamarsh [9]. The conclusion of the theory is that the concentration at a given distance from a stack is a function of the meteorological conditions, the height of the stack, and of course $Q_0$. It was found that for a given height of stack there is a distance at the ground for which we have a maximum concentration. In Fig. 2 we give the distance of maximum concentration as function of the height of the stack. The case presented is for "A" type Pasquill condition [8, 9]. The numbers on the curve give the maximal concentrations in $\mu Ci/cm^3$.

Using now our $Q_0 = 500\ \mu Ci/d$ and wind speed about 1 m/sec we found that for a stack of 120 m heigh the maximum concentration, which corresponds to a distance of 400 m, is about $9 \cdot 10^{-14}\ \mu Ci$ $^{226}Ra$ per $cm^3$ air. This number must be compared to the maximum permissible concentration (MPC) for $^{226}Ra$ in air which is $10^{-11}\ \mu Ci/cm^3$. The concentration found is 2 orders of magnitude lower than the MPC. However since fly ash also contains other radionuclides (e. g. $^{238}U$, $^{210}Pb$, etc.), the problem should be carefully examined with respect to long-term effects on those living close to power plants and especially in the direction of prevailing local winds.

*b) Hazards from wall radioactivity*

In the case that fly ashes should be used in concrete the hazards from direct exposure to its radioactivity can be estimated as follows.

The cement in concrete is about 30% and the proposed substitution is between 20 to 40% [6]. Let us assume 30% fly ashes in the cement. Since 1 $m^3$ of concrete contains about 300 kgr of cement, we found that its activity concentration will be: $P_i = 0.09\ A_i$ where $A_i$ is the activity concentration (number of photons per $cm^3$, emitted in $4\pi$ geometry, per unit time of the particular $i$-th line) of the ash.

Our problem is to calculate the dose from a given energy line at a distance $z$ from a concrete wall containing radioactive fly ash. Firstly we calculate the photon flux at the surface of the wall. This problem is solved in several manuals, e. g. in Price [10].

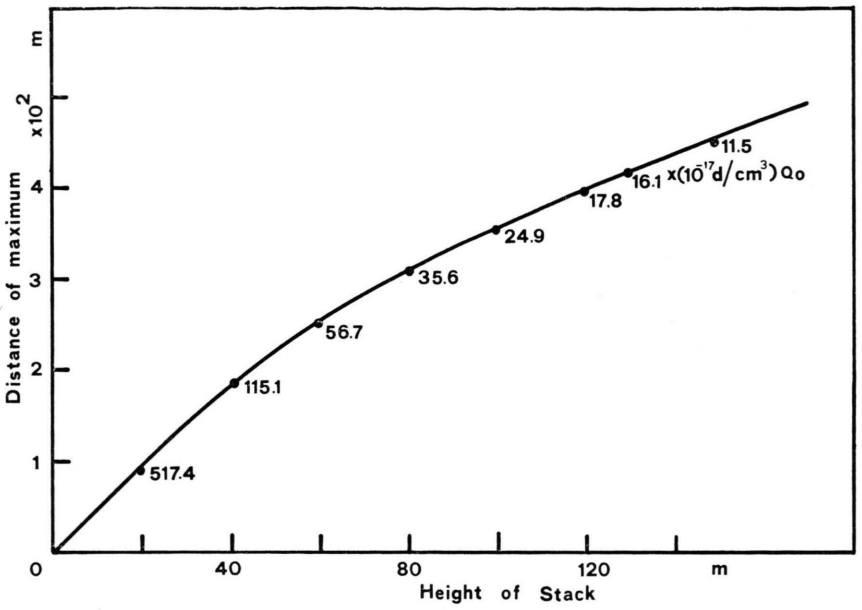


Fig. 2. Distance of maximum radioactive concentration in $\mu$Ci/cm$^3$ as a function of the height of stack.

Next, using the flux of photons so found we calculate the dose from well-known relations or curves.

At a distance of one meter accross a wall of "infinite" area and of "infinite" depth, we estimate about 600 gammas of 609 keV per cm$^2$ and hour. This gives a dose of about $2 \cdot 10^{-4}$ mrad/h of about 2 mrem per year. Assuming that a person stays for 24 hours a day in a room made by concrete of this type we found that he should receive from the 609 keV gammas an annual dose less than 10 mrem per year. By considering all the important gammas of the uranium series in secular equilibrium with the 609 keV gammas and which should be emitted from the "doped" concrete, we calculate an annual dose of the order of 100 mrem. The assumption made in the above estimation was strong, i. e. 24 h permanent living in the active room. So the 100 mrem must be considered as an overestimate.

In practice, if the hazards from the use of radioactive fly ash in concrete should arose only from wall radiation it might be considered as a tolerable risk. This risk, however, should be combined with the hazard from radon diffusing from the wall.

### *c) Hazards from diffusion of Radon through concrete*

If fly ashes should be used in concrete Radon will diffuse out of it. We calculate the hazards from the concentration of Radon in the following manner.

Culot et al. [11] studies the effective diffusion coefficient of Radon in concrete. Applying a linear diffusion theory to diffusion of Radon across a concrete wall they found results indicating that the relaxation distance of Radon in a concrete wall is of the order of 10 cm with an associated effective diffusion coefficent $k_e$ of the order of $2 \cdot 10^{-5}$ cm$^2$/sec. This number is in good agreement with the value of $k_e$ given by Schweite et al. [12].

Using the philosophy of the article of Culot et al., we found that in a room of dimensions $10 \times 10 \times 4$ m$^3$ the concentration of Radon in the air will be about $10^{-9}$ $\mu$Ci/cm$^3$. For the above estimation we used a concrete porosity of 5% and a wall thickness of 20 cm. The concrete composition was taken as in 3 b). The estimated concentration of Radon is about two orders of magnitude lower than that of the MPC of Radon in air, which is about $10^{-7}$ $\mu$Ci/cm$^3$.

In spite of the fact that the hazard from Radon diffused through concrete can be considered as a tolerable one, it may be better however to avoid this doped concrete for construction of buildings for habitation. It is possible to use them for foundations, bridges and other big concrete constructions "en plain air". It must be noted that our estimations for Radon diffusion were made for concrete which has a porosity of 5%, while it is well known that concrete can have porosities of up to 25%. The Radon concentration in air will then be an order of magnitude higher than that given above.

*Appendix*

Average composition of the lignites and fly ashes from the Ptolemais (Kardia) thermopower plant. The weight percentages (Personal communication from CPP, Kardia) correspond to February 1977 and the samples for the radioactive examination were taken from the same material.

| Lignites | | Fly ashes | |
|---|---|---|---|
| Humidity | 56.50 | $SiO_2$ | 30.70 |
| $A_S$hes | 12.80 | $Fe_2O_3$ | 5.58 |
| C | 19.94 | $Al_2O_3$ | 14.56 |
| $H_2$ | 1.40 | $TiO_2$ | 0.22 |
| S | 0.53 | CaO | 34.67 |
| $N_2$ | 0.38 | MgO | 3.47 |
| $O_2$ | 8.45 | $SO_3$ | 8.47 |
| | | $Na_2O$ | 0.53 |
| | | $K_20$ | 0.88 |

[1] Z. Jaworowski, J. Bilkiewicz, and E. Zylicz, Health Phys. **20,** 449 (1971).
[2] W. Anderson and R. Turner, Nature (Lond.) **178,** 203 (1956).
[3] W. Anderson, W. Mayneord, and R. Turner, Nature (Lond.) **174,** 424 (1954).
[4] H. Kirchner, E. Merz, and A. Schiffters, Braunkohle **26,** 340 (1974).
[5] M. Venuat, Rev. Mater. Constr. **692,** 30 (1975).
[6] E. Voyatzakis, K. Sipitanos, and A. Christaki-Papageorgiou, Rev. Mater. Constr. **703,** 341 (1976).
[7] C. Papastefanou and St. Charalambous, Nucl. Instrum. Meth. **151,** 599 (1978).
[8] F. Pasquill, Atmospheric Diffusion, Van Nostrand, Amsterdam 1962.
[9] J. Lamarsh, Introduction to Nuclear Engineering, Addison-Wesley, London 1975.
[10] B. Price, C. Horton, and K. Spinney, Radiation Shielding, Pergamon Press, London 1957.
[11] M. Culot, H. Olson, and K. Schiager, Health Phys. **30,** 263 (1976).
[12] H. Schweite, H. Bohone, and U. Lugwing, Proc. 5th Int. Symp. on the Chemistry of Cement Japan (1968).