

## HAZARDS FROM RADIOACTIVITY OF FLY ASH OF GREEK COAL POWER PLANTS (CPP)

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**Abstract.** Fly ash and fine dispersion releases by coal combustion in Greek CPPs are radioactive. Concentrations in the fly ash up to 20 pCi/g and 10 pCi/g were measured for  $^{238}\text{U}$  and  $^{226}\text{Ra}$  respectively (not in secular equilibrium). The radioactivity of fly ash deduces risks in two ways: a) from the escaping fly ash in particulate form or fine dispersion and b) from using fly ash as substitute of cement in concrete.

**1. Introduction.** Coal burning by CPP is one of the principal sources of Radioactivity of the Atmosphere (1-8). The radioactivity escapes from stacks of CPPs in particulate form (fly ash) or fine dispersion. In both cases a potential hazard in the vicinity of CPPs is constituted.

It has been proposed (9-12) to use fly ash as substitute of cement in concrete. Since fly ash would be radioactive, its use could involve potential hazards, arising, either from direct irradiation from concrete or from radon diffusion from it.

**2. Experimental Procedures and Results.** The samples of fly ash studied were from the Greek CPP's. Their radioactivity was mainly measured by Ge-Li detectors using the direct  $\gamma$ -spectroscopy and activation analysis by thermal neutrons. Fig. 1 shows a typical gamma spectrum. In the spectrum we clearly see the important gamma transitions of the uranium series from  $^{226}\text{Ra}$  onwards. The  $\gamma$ -peaks of the thorium series and of  $^{40}\text{K}$  are absent. The results for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  are presented in Table 1a. In 1b, the concentrations of the above nuclides for the lignites of principal Greek Coal Mines, burning in the Greek CPP's, are also presented. It was found that uranium and radium are not in secular equilibrium in lignites and fly ashes, but the physics involved escapes of the present paper.

**3a. Radioactivity escaping in particulate form of fly ash.** From the stacks of CPP, fly ash escapes in a percentage of 3% or 1% of the total fly ash, in the better cases. The total amount escaping per each Greek CPP reaches up to about  $7.5 \times 10^{11} \text{ g/y}$ . By differential sieves and weighing, the grain distribution of fly ash was determined. In the Fig. 2, the distribution function of fly ash consisted of grains with diameter ranged between  $\delta$  and  $\delta + \Delta\delta$  is presented. We observe that this distribution presents a strong maximum of grain mass with mean diameter ranged between 20 and 70  $\mu$ .

The horizontal distance  $x$ , in meters, in which the grain of mean diameter  $\delta$  will fall on the ground surface from the height  $h$  of the stack, is given by the equation, derived by Stokes law:  
$$x = 1.36 \times 10^4 \cdot (h/\delta^2) u^2 \text{ (meters)},$$
 where  $h$  the height of the stack, in meters,  $u$  the mean wind speed in m/sec and  $\delta$  the mean grain diameter in  $\mu$ . As a consequence of the distribution function of Fig. 2, the deposition of fly ash shows a maximum which, for the case of  $u=1\text{m/sec}$ , is at a

region about 400 m upwind of the stack, Fig.3. This maximum is removed to major distances as much as the wind speed is increased. On the site of maximum,  $^{226}\text{Ra}$  deposition has the value:  $6.2 \times 10^{-9} \mu\text{Ci}/\text{cm}^2 \cdot \text{d}$ .

3b. Radioactivity escaping from fine dispersion. Taking in consideration the quantity of coal burning per each CPP, which is about  $7.3 \times 10^{12} \text{ g/y}$ , the ash content (12%), fly ash release (80% of the total ash content), as well their  $^{226}\text{Ra}$  specific activities, it is deduced that  $42 \text{ Ci/y}$   $^{226}\text{Ra}$  is escaping as fine dispersion (finest particulate form or maybe in gaseous form) from a stack of CPP.

#### 4. Hazards from the Radioactivity escaping from CPP.

a) Hazards from the fly ash. In the site of maximum, for  $u=1 \text{ m/sec}$ , we can find a  $^{226}\text{Ra}$  concentration in the air about  $2.3 \times 10^{-9} \mu\text{Ci}/\text{cm}^3$ . Also, if we regard all the radio-elements of the uranium series in the grains of fly ash, as well preferable wind direction, we shall have again that the total radioactive concentration, arising from fly ash, will be 20 times lower than the MPC ( $\sim 10^{-11} \mu\text{Ci}/\text{cm}^3$ ).

b) Hazards from fine dispersion. In Fig.4 typical curves are presented that they give the maximum concentration,  $x_{\text{max}}$ , in several distances from the stack, horizontally, on its base, in the main direction of the local wind blown, as a function of the height of the stack. Each curve corresponds to one Pasquill condition (13), from A (extremely unstable) to F (extremely stable). For our calculations (14) a wind speed of  $1 \text{ m/sec}$  was used. In the case of a radioactive release (CPP Kardias-Ptolemais)  $Q_0=42 \text{ Ci/y}$  of  $^{226}\text{Ra}$ , it is found that  $x_{\text{max}}$  is about  $2.10^{-11} \mu\text{Ci}/\text{cm}^3$  (Pasquill condition "A"). This is the same value as the MPC in the air. The above consideration had as presupposition a stable and singular wind direction. This hypothesis is non-realistic and therefore the maximum concentration of  $^{226}\text{Ra}$  will be lower than the one estimated. However, other toxic radionuclides escape from the stack of CPP such as:  $^{238}\text{U}$ ,  $^{210}\text{Pb}$ , etc., which leads to hazard in other (bad) directions.

For the most common Pasquill condition F (extremely stable) the whole body man dose exposure at the distance of 400 m from a stack 120 m in height and  $u=1 \text{ m/sec}$ , was calculated. Using the philosophy of calculations as is given by Cohen et al (15), it was found that for  $42 \text{ Ci/y}$  of  $^{226}\text{Ra}$  this is about 0.5 man-rem. This must be compared with 3 man-rem which is the permissible whole body man dose exposure.

#### 5. Hazards from use of fly ash as substitute of cement in concrete.

a) Hazards from wall radioactivity. The cement in concrete is about 30% and the proposed substitution is between 20 to 40% (16). Let us assume 30% fly ashes in the cement. At a distance of one meter across a "doped" wall of "infinite" area and of "infinite" depth, the calculations give 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.

#### b. Hazards from diffusion of Radon through concrete.

Radon is diffused across a doped concrete wall. Culot et al (17) studied extensively the problem. Using the philosophy of Culot et al, we found that in a room of dimensions  $10 \times 10 \times 4 \text{ m}^3$  the concentration of Radon in the air will be about  $10^{-9} \mu\text{Ci}/\text{cm}^3$ . For the above estimation we used a concrete porosity of 5% and a wall thickness of 20 cm. The estimated concentration of Radon is about two orders of magnitude lower than that of the MPC of Radon in the air, which is about  $10^{-9} \mu\text{Ci}/\text{cm}^3$ . However, if we used a 25% porosity, the Radon concentration will be an order of magnitude higher.

6. Conclusions. From the experimental data and discussion we can conclude: a) The major component of the risk is due to the fine structure of release. b) Fly ash would be avoided for use in doped concrete for habitation. It would be used for other concrete constructions "en plain air".

It is expected that the coal use for electric power generation to be increased by a factor almost 10 in the next 10 years. Several estimations, i.e. Klein et al (18), Bertine and Goldberg (3) and Ondov et al (19), are made on the atmospheric releases of various potentially toxic elements from large CPP. In them, the release of radioactive elements must be added. All of them must be taken

seriously in consideration in the designing of new large CPP's, since as the present work demonstrates, could have concentrations of radioactivity close, as well greater, to MPC.

The new data indicate, as Eisenbud and Petrow (1), Kolb (20) and Auran (21) estimate, that the Coal Power Plants discharge relatively larger quantities of radioactive materials into the atmosphere than Nuclear Power Plants, of comparable size, during their "normal" operation.

#### References

1. M.Eisenbud and H. Petrow, Science 144(1964)288.
2. E.J.Martin, D.E. Harward, M.J.Smith and H.P.Bedrosian, CONF-700-810-24 (1969) USAEC, Oak Ridge, TN.
3. K.K.Bertine and E.D.Goldberg, Science 173 (1971) 233.
4. Z. Jaworowski, J. Bilkiewicz and E.Z.Zylicz, Health Phys.20(1971)449.
5. H.Moore and S. Poet, Atmos.Env. 10(1976)381.
6. D.G.Coles, R.C.Ragaini and J.M.Ondov, Environ.Sci.Technol.12(1978)442.
7. J.P.Mc Bride, R.E.Moore, J.P.Witherspoon and R.E.Blanco, Science 202(1978)1045.
8. C.Papastefanou and S.Charalambous, Z. Naturforsch. 34a(1979)533.
9. M.Vennat Rev.Mater. Constr. No 692(1975)30.
10. B.Zlatanov "2<sup>e</sup> Congrès Grec sur le béton armé"Thessalonique,Grèce 28-31 Mai 1975.
11. K.Sipitanos, E.Voyatzakis and S.Melidis Rev.Mater. Constr. No 707(1977)211.
12. I.Papayanni-Papadopoulou "3<sup>e</sup> Congrès Grec sur le béton armé" Chios, Grèce 1-4 Octobre 1977.
13. F. Pasquill, Atmospheric Diffusion, Van Nostrand, Amsterdam 1962.
14. J.Lamarsh, Introduction to Nuclear Engineering, Addison-Wesley, London 1975.

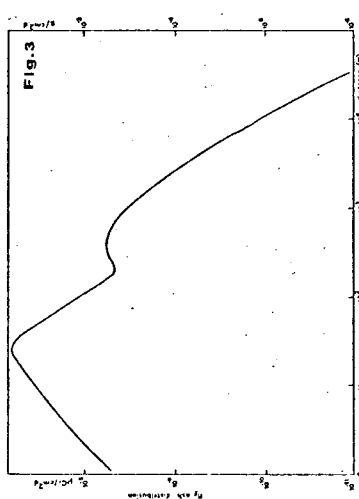
15. B.L.Coben, H.N.Jow and I.S.Lee, Health Phys. 34(1978)569.
16. E.Voyatzakis, K.Sipitanos and A.Christaki-Papageorgiou, Rev.Mater Constr. 703(1976)341.
17. M.Culot, H.Olson and K.Schiager, Health Phys. 30(1976)263.
18. D.H.Klein, A.W.Andren, J.A.Carter, J.F.Emery, C.Feldman, W.Fulkerson, W.S.Lyon, J.C.Ogle, Y.Talm, R.I.Van Hook and N.Bolton, Environ. Sci. Technol. 9(1975)973.
19. J.M.Ondov, R.C.Ragaini and A.H.Bierman CONF-771072(1977) p.338-357.
20. W.Kolb, PTB-Ra-8 Feb.1978 13p.
21. K.Aurand, Wiss. Umwelt, ISU (1978) (no.2) p.65-74.

Table 1. a) Radioactivity of Fly Ashes from Greek CPP

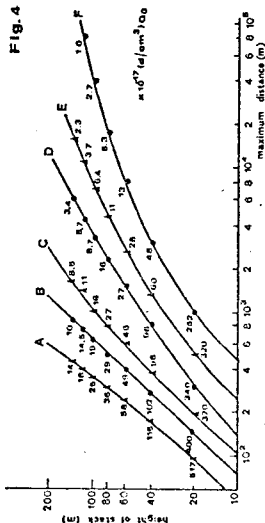
CPP	$^{235}\text{U}$ pCi/g	$^{210}\text{Ra}$ pCi/g	$^{232}\text{Th}$ pCi/g
Kardia Protemais	23.2±3.3	10.4±0.9	0.192±0.014
Aliveri	14.3±3.3	8.3±0.9	0.035±0.012
Megalopolis	13.3±3.3	10.6±0.9	0.193±0.026

#### b) Radioactivity of Greek Lignites

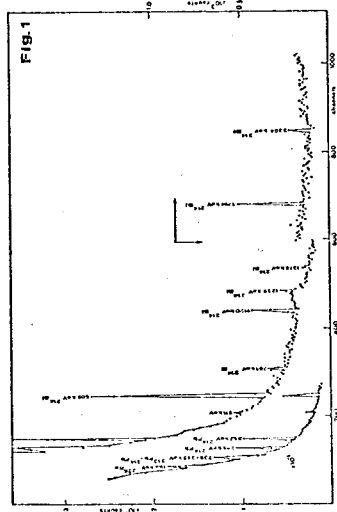
Coal Mine	$^{238}\text{U}$ pCi/g	$^{226}\text{Ra}$ pCi/g	$^{232}\text{Th}$ pCi/g
Mardia Ptolemais	11.4±3.4	6.5±3.8	3.024±0.005
Aliveri	8.0±3.4	3.6±0.7	0.019±0.037
Megalopolis	11.7±3.4	3.2±0.7	0.023±0.005
Serres	35.3±5.4	11.2±2.1	0.021±0.003
Vevi	—	8.4±0.8	0.024±0.004



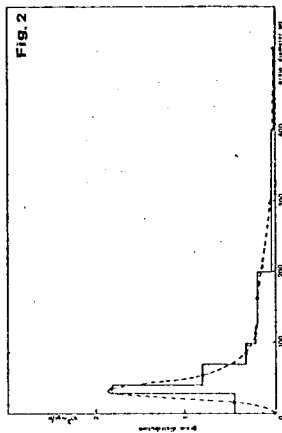
3. Distribution of fly ash and its radioactivity depositing in one direction (wind blown) from the stack released as a function of the distance downwind.



4. Typical curves for various Pasquill conditions A to F. The numbers on the curves show the maximum concentration of Ra-226 ( $\mu\text{Ci}/\text{cm}^3$ ) at several distances downwind from the stack released as a function of the height of the stack.  $Q_0$  is the radioactive release in  $\mu\text{Ci}/\text{d}$ .



1. Typical "net" gamma spectrum of fly ash from Kardias Ptolemais. The Background is subtracted.



2. Histogram of distribution of particle size of fly ash ( $g/\mu$ ) as a function of the mean diameter ( $\mu$ ) of grains.