

TECHNOLOGICALLY ENHANCED NATURAL RADIOACTIVITY OF FLY-ASH PRODUCED IN LIGNITE-FIRED POWER PLANTS AND THE ASSOCIATED RADIOLOGICAL IMPACT

D.J. Karangelos, P.K.Rouni, N.P. Petropoulos, M.J. Anagnostakis, E.P. Hinis and S.E. Simopoulos

Nuclear Engineering Section, Mechanical Engineering Department
National Technical University of Athens, 15780 Athens, Greece

1 ABSTRACT

Coal- and lignite-fired power plants produce significant amounts of ashes, which are quite often being used as additives in cement and other building materials. In many cases, coal and lignite present high concentrations of naturally occurring radionuclides, such as ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th and ^{40}K . Moreover, during the combustion process the produced ashes are enriched in the above radionuclides, thus characterized as *Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM)*. The most important naturally occurring radionuclide in such power plant ashes is ^{226}Ra , since it produces radon, which when exhaled contributes significantly to the dose received by workers and the public. An extensive research project for the determination of the natural radioactivity of lignite and ashes from Greek lignite-fired power plants is in progress in the Nuclear Engineering Section of the National Technical University of Athens (NES-NTUA) since 1983. From the results obtained so far, it may be concluded that ^{226}Ra radioactivity of fly-ash in some cases exceeds 1 kBqkg^{-1} , which is very high compared to the mean ^{226}Ra radioactivity of surface soils in Greece (25 Bqkg^{-1}). Furthermore, the radioactivity of ^{210}Pb in fly-ash reaches up to 4 kBqkg^{-1} , depending on the sampling location inside the power plant. This paper presents: natural radioactivity results from the analysis of the lignite fed to and the ashes produced in Greek lignite-fired power plants, and enrichment ratios of natural radionuclides in the ashes collected at different stages along the emission control system of the power plant. Enrichment ratio results are being interpreted in relation to the physical properties of the investigated nuclides and the temperature in the flue gas pathway. Discarded ash deposition fields in the vicinity of the power plants are also investigated in terms of gamma-ray dose rate, soil gas radon concentration and surface soil radon exhalation.

2 INTRODUCTION

Fossil fuels such as coal and lignite play an important role within the electric power generation system worldwide. Coal, lignite and their combustion residues (fly-ash and bottom-ash or slag) contain trace elements such as As, Sb, Cd, Cr,

Pb, Ni, Se, Zn and all the *Naturally Occurring Radionuclides (NOR)*, among which the most important are: ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Ra and ^{40}K . According to [Zielinski R.A., 1998], modern power plants, burning typical feed coals produce a barely perceptible (1-5%) increase in the radiation dose to the public, above that due to the natural background radiation. The above conclusion is based upon the assumption that the fossil fuels used do not have unusually high radioactivity, and that the efficiency of the emission control devices of the power plants is high (> 99.5%). Since this is not always the case, a systematic research has to be carried out in order to verify the above conclusion. Even with the use of high efficiency emission control devices, other problems, such as radon exhalation, and the leachability of the natural radionuclides in the large volumes of recovered coal combustion residues still exist [Zielinski R.A., 1998]. Combustion residues are either permanently deposited, or temporarily stored at the plant sites destined for commercial utilization. In both cases natural radionuclides existing in the ashes may be leached and enter the terrestrial or aquatic environment.

In Greece, almost 80% of the electric power is produced in lignite-burning thermal power plants. Today, there are two very important lignite deposits under exploitation. The first is the Ptolemais deposit, in the Northern part of the country, which, from various fields, supplies local power stations with a total installed capacity of about 3700MW. The second is in the South of Greece, near the small town of Megalopolis, feeding power plants with total installed capacity of 900MW. Lignite of both deposits, and especially that of the Megalopolis deposit, is of low calorific value, has rather high water and ash content and is amongst the poorest used for electricity generation. Furthermore, in both deposits, and especially in that of Megalopolis, the natural radioactivity of lignite is much higher than that of the surrounding soil [Simopoulos S.E., 1987], due, presumably, to the leaching from uraniferous rocks in the vicinity. From this point of view these lignites have to be considered of high radiological importance.

During the combustion process in the power plant, the removal of all combustible elements results to a significant enrichment of all incombustible elements in the produced ash, among which are all the natural radionuclides, thus characterizing the ashes as *Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM)*. Furthermore, due to the different physicochemical properties of the various incombustible elements in the fuel, the enrichment of the various trace elements may differ in the various partitions of the collected ashes. The incombustible elements entering the boiler in the coal stream are partitioned between the bottom-ash (or slag), which falls inside the boiler, and the fly-ash, which is suspended in the flue-gas together with vapours of volatile elements and compounds. A further partitioning of ash in the flue-gas stream takes place in the particulate emission control devices – and especially in the electrostatic precipitators (ESP) - that efficiently remove larger fly-ash particles, but are less efficient for vapours and finer particles.

In this paper presents recent results of the research which is being conducted in the Nuclear Engineering Section of the National Technical University of Athens (NES-NTUA), on the radiological characteristics of the ashes produced in greek

lignite burning power plants and the subsequent radiological consequences from lignite burning in Greece.

3 NATURAL RADIOACTIVITY OF GREEK LIGNITES AND ASHES.

3.1 Sampling and analysis

The investigation of the natural radioactivity of greek lignites and ashes in NES-NTUA started back in 1983. Samplings and analyses have been performed since then in many lignite-mines allover Greece as well as in lignite fired power plants. For each power plant investigated the following sampling schemes have been conducted [Simopoulos S.E., 1987]:

- Sampling of lignite in all the lignite fields feeding the power plants. Lignite samples were collected from each lignite seam and intermediate layers of the lignite mine, to allow for the investigation of the natural radioactivity variability in the lignite feeding the power plant.
- Daily sampling of lignite feeding the boiler in the power plant and the fly-ash and the bottom-ash produced. For each power plant, this sampling lasted for five consecutive weeks. The collected samples were used to prepare five weekly representative specimens for lignite, fly-ash and bottom ash.
- In the case of the Megalopolis-IV power plant, fly-ash was systematically collected from various points along the emission control system of the plant, within a period of about three weeks.

During the sampling period the output of the units was kept as constant as possible.

The collected samples were analysed using gamma spectroscopic techniques for the determination of the natural radionuclides: ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Ra and ^{40}K . For the γ -spectroscopic determination of the above radionuclides high resolution high efficiency Ge detector set-ups, and detectors such as a LEGe and XtRa were used. Details about the techniques used may be found elsewhere [Simopoulos S.E., 1987]. ^{226}Ra activity was determined both: indirectly from its decay products in equilibrium and directly from the 186.25 keV photons, in the cases where the analysis of the multiplet photopeak at ~186keV was applicable [Petropoulos et al, 2001]. In the cases where low energy photons (below 200keV) were used for the radionuclide determination, such as ^{238}U (63.29keV) and ^{210}Pb (46.54keV), efficiency correction techniques were used to take into account for the self-absorption of the low energy photons inside the samples analysed [Anagnostakis M.J., 1995].

3.2 Technologically Enhanced Natural radioactivity of ashes produced in greek lignite fired power plants.

Due to the organic matter burn-out during the combustion process, the concentration (ppm) of all the trace elements remaining in the produced ashes, among which are all the natural radionuclides, increases, depending of course on the physicochemical properties of the specific trace element. In the case of the nuclides of the uranium series, the three most important nuclides: ^{238}U , ^{226}Ra and ^{210}Pb have different physicochemical properties, resulting in a different behavior inside the power plant. According to [Coles D.G., 1978], the behavior of uranium during the combustion process depends upon the conditions of the furnace as well as its chemical and physical characteristics of the input fuel. For example, since uranium may exist in the fuel both as uraninite and coffinite, volatile and nonvolatile species may be formed during combustion. ^{226}Ra behaves in a similar way. A portion of ^{226}Ra will reside with the uraninite fraction of its ^{238}U parent, allowing for more mobile species, than the silica associated ^{226}Ra . ^{210}Pb which is more volatile leaves the boiler with the flue-gas in gaseous form and condenses as the temperature of the flue-gas drops.

The results of the analysis of lignite and the ashes produced in thermal power plants from all over the world are presented in Table 1. From this table it is concluded that:

- Greek lignite and ashes have relatively high levels of natural radioactivity, with the Megalopolis lignite having the higher levels of natural radioactivity, among the lignites used today in Greece.
- ^{238}U and ^{226}Ra enrichment in fly-ash is higher than that in bottom-ash.
- ^{210}Pb is highly enriched in fly ash, while in bottom ash it is in some cases depleted.
- The difference of the enrichment of the ^{238}U , ^{226}Ra and ^{210}Pb in the ashes, result in radioactive disequilibrium in the ashes as described by [Coles D.G, 1978]. It should be noticed that, as verified from the analyses performed in NES-NTUA [Anagnostakis,1998], radioactive equilibrium among the nuclides of the uranium series is quite often observed in the lignite feeding greek power plants, though cases of disequilibrium are also reported [Papastefanou C., 1982].

In order to further investigate the natural radioactivity of the fly-ashes collected inside the emission control system of the power plant Megalopolis-IV, a detailed sampling inside the power plant was performed. Fly-ash samples were periodically collected at: the water preheater (Economizer), the air preheater (LUVVO) and the four rows in each of the two lines (right and left) of the Electrostatic Precipitators (ESP), along the emission control system of the plant. In Figure 1 are presented the sampling points inside the power plant, together with estimated flue gas temperatures and the percentage of ash collected in each point. Again, the collected samples were analysed using gamma-spectroscopic techniques.

The activities (range, mean value and standard deviation) of ^{238}U , ^{226}Ra , ^{210}Pb and ^{40}K in the fly-ash samples collected in each one of the 10 sampling points, within the period of 3 weeks are presented in Table 2. The cumulative results of

the activity of ^{238}U , ^{226}Ra , ^{210}Pb and ^{40}K , of the samples collected: at the Economiser, the Luvo and the four rows of the left line of ESP, are presented in Figure 2. From the results presented in Table 2 and Figure 2 it may be concluded that:

- a. The natural radioactivity of the fly-ash produced in the power plant presents significant variations over time, depending on the activity of the feeding lignite, even for the relatively small period of 3 weeks. For this reason care should be taken while sampling in the power plant in order to obtain representative results.
- b. The natural radioactivity of the fly-ash significantly depends on the sampling location, due mainly to the different temperatures of the flue gas, as well as the grain size distribution of the fly-ash collected in each point.
- c. ^{226}Ra activity in the fly-ash does not significantly change within the emission control system.
- d. ^{238}U activity in the fly-ash slightly increases towards the end of the emission control system. The characteristics and the behaviour of uranium during the combustion process depend upon by the conditions inside the furnace as well as its chemical and physical form in the input lignite.
- e. ^{210}Pb , the most volatile among the natural radionuclides in the fly-ash, is more enriched in the coldest parts of the emission control system, where the finest particles of the fly-ash are collected [Coles D.G, 1978], [Anagnostakis,1998]. During the combustion process, ^{210}Pb is removed from the furnace together with the flue gases in gaseous form, and as the temperature of the flue-gases and the fly-ash is reduced, ^{210}Pb condenses out, preferentially on the finer fly-ash particles, which are cooled first. These finer fly-ash particles have higher surface-to-volume ratio, resulting to higher specific activity of the condensed ^{210}Pb .
- f. ^{40}K activity does not change significantly along the emission control system.

In order to statistically justify the significance of the ^{210}Pb activity differences, along the emission control system, a Tukey statistical test was performed. Since for some points very few samples were analysed (e.g. for the 4th row of ESP, samples for only two samples were analysed), the whole Tukey test was based on the analysis of two fly-ash samples, each collected for a three hours period, for every sampling point. From the results of the test it was concluded that:

- a. The ^{210}Pb activity of fly-ash collected along the emission control system depends on the sampling point.
- b. The ^{210}Pb activity of the fly-ash samples collected in the LUVO, Economizer and the 1st row of ESP, does not significantly differ.
- c. The ^{210}Pb activity of the fly-ash samples collected in the 3rd and the 4th row of ESP, does not significantly differ.

The analysis of a few more samples from the last rows of ESP is needed to improve the justification of the argument that ^{210}Pb activity of fly-ash collected along the emission control system depends on the sampling point.

Due to the different enrichment of the natural radionuclides ^{238}U , ^{226}Ra and ^{210}Pb along the emission control system, and given the fact that radioactive equilibrium exists in the lignite feeding the power plant, a disturbance of the

radioactive equilibrium among the above nuclides in the produced ashes is expected.

The ratios of the activities of $^{210}\text{Pb}/^{226}\text{Ra}$ for all the samples collected along the emission control system are presented in Table 3, while in Table 4 presents the mean ratios of $^{238}\text{U}/^{226}\text{Ra}$ and $^{210}\text{Pb}/^{226}\text{Ra}$, for each sampling point.

The estimated total uncertainty of these ratios is less than 10%.

From Tables 3 and 4 may be concluded that:

- i. Due to the different enrichment of ^{238}U , ^{226}Ra and ^{210}Pb , there is a significant disruption of radioactive equilibrium in uranium series.
- ii. Regardless of the fluctuations of fly-ash activity, the ratios of the above nuclides at the sampling points remain more or less constant with time.
- iii. In the Economizer and the LUVO, which are hottest parts of the emission control system, ^{210}Pb is depleted compared to ^{226}Ra and ^{238}U , with the ratio $^{210}\text{Pb}/^{226}\text{Ra}$ reaching the value of 0.6.
- iv. In the 4th row of the ESP, which is the coldest part of the emission control system, ^{210}Pb is highly enriched compared to ^{226}Ra and ^{238}U , with the ratio $^{210}\text{Pb}/^{226}\text{Ra}$ reaching the value of 3.5.
- v. The ratio $^{238}\text{U}/^{226}\text{Ra}$, for both lines of ESP, is less than 1 in the hottest side of the emission control system, and higher than 1 in the coldest side.
- vi. ^{210}Pb activity in the fly-ash collected in the fourth line of the ESP parts may be up to 6 times higher than in the fly-ash collected at LUVO.

It should be noted that natural radionuclides enrichment and dependence upon the sampling location inside the power plant has also been reported for other fuels such as oil-shales [Realo K., 1997].

Regarding the radioactivity of the escaping fly-ash, one sample of fly-ash that was collected inside the stack of another power plant from the Ptolemais region and analysed, showed that the ^{226}Ra radioactivity of the escaping fly-ash did not significantly differ from that collected at the ESP. However, further investigation for other isotopes as well is needed.

4 EXHALATION MEASUREMENTS IN DISCARDED FLY-ASH DEPOSITION FIELDS.

In order to investigate the radioenvironmental impact due to discarded fly-ash from the power plants, a series of measurements were performed at discarded fly-ash deposition fields, in the vicinity of the power plants of Megalopolis. The measurements conducted were: radon in ambient air concentration, radon exhalation from the ground, soil gas radon concentration, and external dose-rate due to gamma radiation 1m above ground surface. Three deposition fields were chosen: two fields that were restored and planted during mid 70's and mid 80's respectively, and one exhausted open lignite mine, currently being used as deposition field, which has not been restored yet. For comparison purposes similar measurements were also performed in an undisturbed nearby field, where presumably lignite deposits exist, that was chosen as a reference point. From Table 5, where the results of the above measurements are presented, it is concluded that:

- Gamma-ray dose rate is in all cases elevated, compared with that due to natural background in Greece (46nGyh^{-1}), including in the reference point. The highest dose rate was observed in the deposition field that is not restored yet (500nGyh^{-1}). This result was expected, since the soil on the restored fields shields gamma-rays emitted by the fly-ash.
- Radon concentration in the ambient air, in soil gas, and radon exhalation from the ground have low to medium values, in all deposition fields. Surprisingly, radon concentration in soil gas and radon exhalation from the ground are much higher in the reference point. One possible explanation for these results is that the emanation of radon from the fly-ash grains is low, because of the crystallisation of the grain surface, during the combustion process, thus leading to very low exhalation rate from fly-ash deposits, though ^{226}Ra content of the fly-ash is high. This crystallisation highly depends on the silica content of the lignite, as well as the temperatures inside the furnace. It should be noted that the fly-ash from the Megalopolis region is rich in quartz and aluminosilicates [Sakorafa V. et al, 1996]. The high ^{226}Ra content of soil gas, should be attributed to the presumably existing lignite deposits.

Further investigation is underway in order to determine the radon exhalation rate from the fly-ash samples collected along the emission control system of Megalopolis-IV power plant. A more detailed investigation of the fly-ash deposition fields is also underway.

5 CONCLUSIONS

Ashes produced in thermal power plants may contain high levels of natural radioactivity and constitute a potential health hazard to the power plant personnel, and to the population living in the vicinity, due to fly-ash releases, fly-ash depositions and fly-ash industrial utilization. In order to determine the potential risk due to fly-ashes, a detailed investigation inside the power plant should be conducted. The radioactivity of the produced ashes highly depends on the point where the ash is collected inside the power plant, while radioactive equilibrium among the nuclides of ^{238}U series is quite often significantly disturbed in the produced ashes. In the case of ^{210}Pb , its activity in fly-ash may vary up to 6 times, depending on the sampling location inside the power plant. Thus, when fly-ash is to be collected from the emission control system, in order to be further utilized in other industrial processes, a careful selection of the fly-ash collection point may result to the reduction of the possible health risk due to the fly-ash radiological characteristics, provided that the physicochemical characteristics of this ash allow for the specific use.

The external gamma-ray dose rate in discarded fly-ash deposition fields may be much higher than that of the natural background. Though ^{226}Ra activity of the deposited fly-ash may be very high, exceeding in some cases the value of 1kBqkg^{-1} , and leading to an increased gamma-ray dose rate, in the case of the Megalopolis lignites, the radon exhalation from discarded fly-ash fields is not necessarily high, depending on the lignite origin and the conditions inside the boiler.

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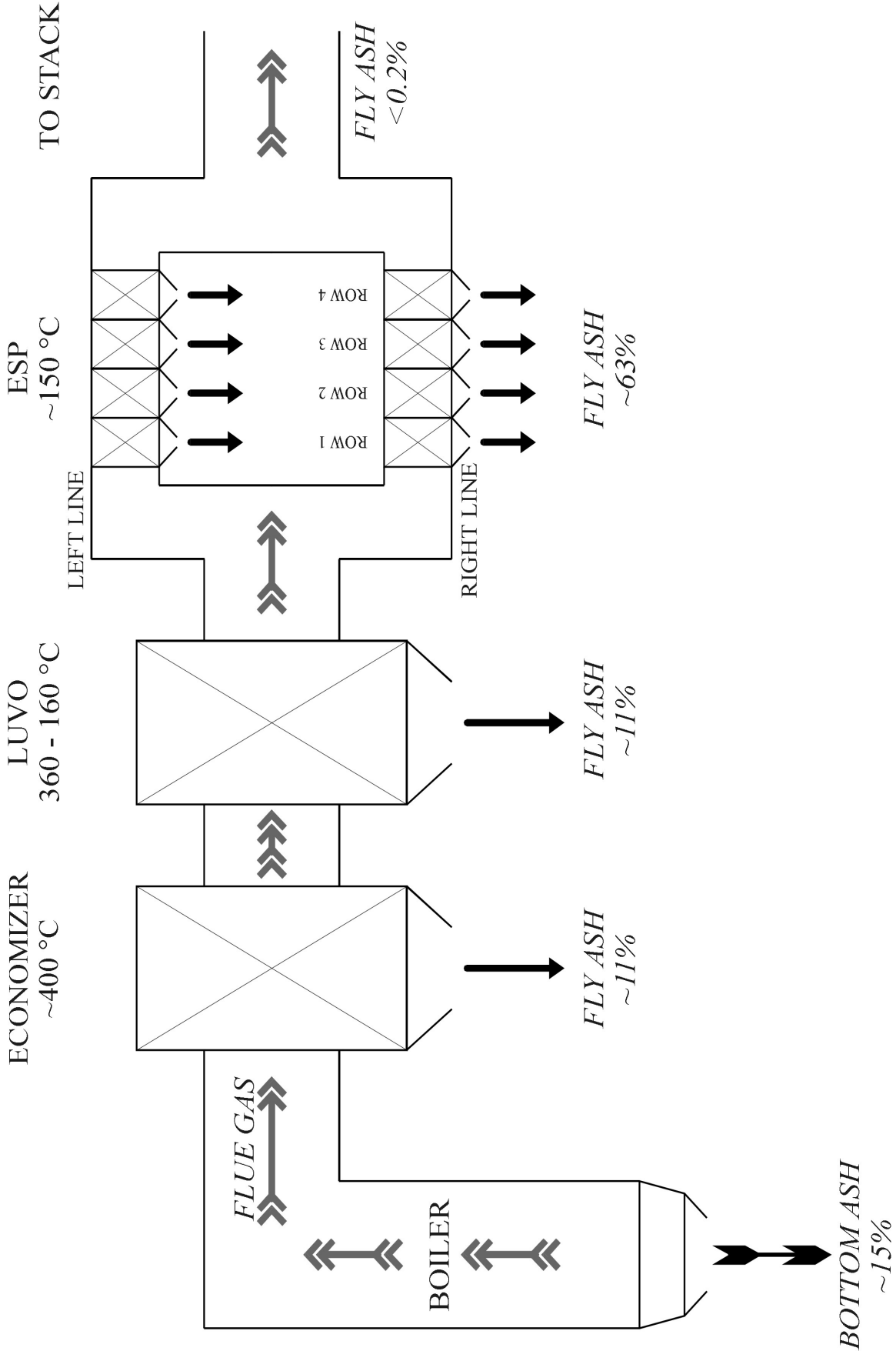


Figure 1. Ash discharges from Megalopolis-IV lignite fired power plant

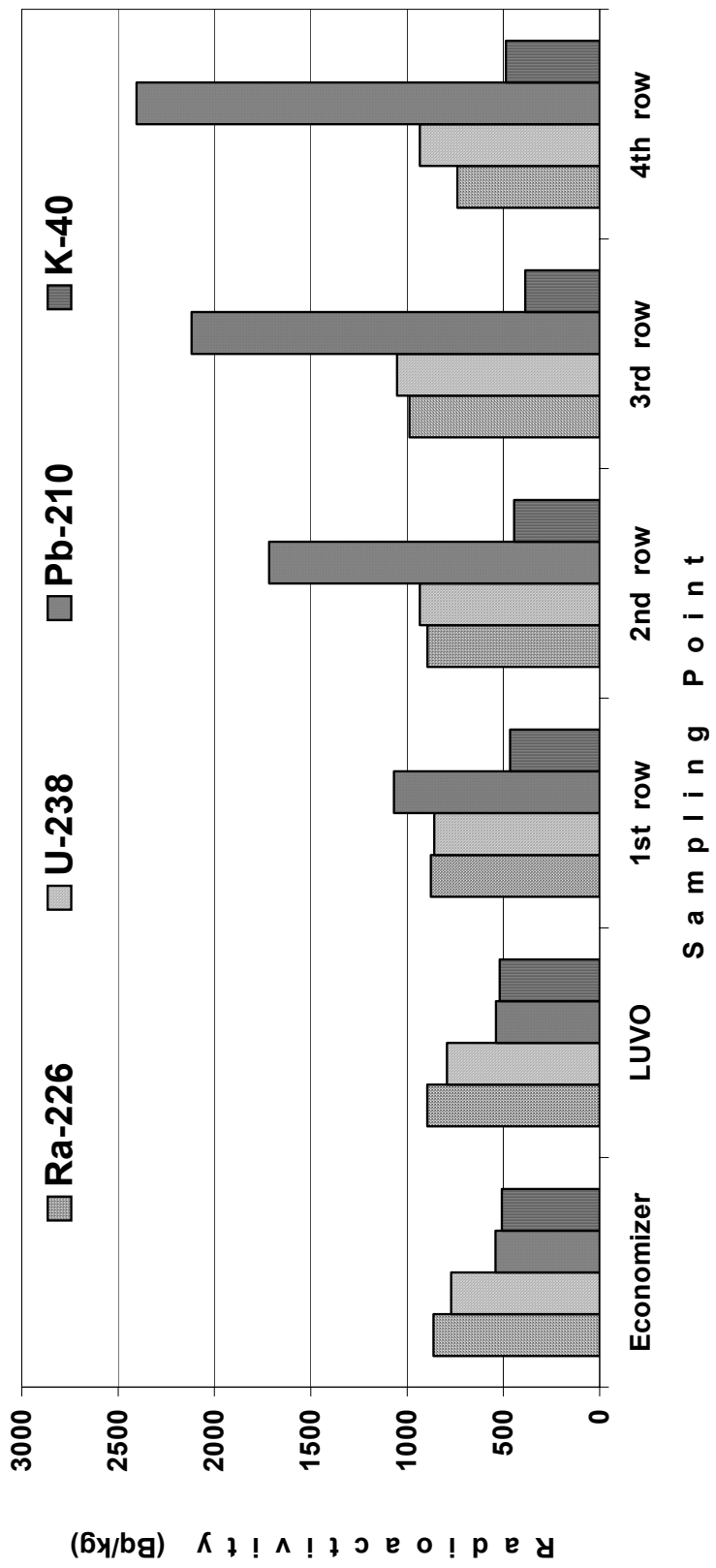


Figure 2. Natural radioactivity of fly-ash collected in ECONOMIZER, LUVO and the left line of ESP of Megalopolis-IV power plant

Table 1. Natural radioactivity of lignite feeding and the ashes produced in Power Plants worldwide.

20. 21.	Power Year unit	Lignite (Bqkg ⁻¹)					Fly-ash (Bqkg ⁻¹)					Bottom-ash (Bqkg ⁻¹)					Ref.
		²³⁸ U	²²⁶ Ra	²¹⁰ Pb	²²⁸ Ra	⁴⁰ K	²³⁸ U	²²⁶ Ra	²¹⁰ Pb	²²⁸ Ra	⁴⁰ K	²³⁸ U	²²⁶ Ra	²¹⁰ Pb	²²⁸ Ra	⁴⁰ K	
	Megalopolis Unit I	1985	314±52		21±2	181±26	807±38		55±2	449±47		546±77		44±3	406±63	[17]	
	Megalopolis Unit III	1985	355±13	321±40	370±13	21±1	928±9.2	845±71	948±9.5	502±21	590±11	587±71	372±13	44±2	423±30	[4], [17]	
	Megalopolis Unit IV	1997	306±13	346±8.2	361±9.6	19±8.5	964±6.6	904±8.6	1158±11	454±11	681±3.5	662±9.3	275±5.9	41±5.0	405±11	[4]	
	Ptolemais Unit IV	1986		83±6		78±13	261±19			251±16		114±16		24±3	137±21	[16]	
	Kardia Unit I	1986		175±20		68±13	600±75		49±5	217±17		363±29		36±2	200±11	[16]	
	Kardia	1979	126±37	255±30			260±37	385±33			144±37	174±33				[9]	
	Kardia	1984					340-640	190-500								[11]	
	Megalopolis	1984						590-850	58-80							[7]	
	Kardia	1979	388-422	255±30			503±851	385-421								[9]	
	Megalopolis	1979	433±126	118±26			492±122	392±33								[9]	
	USA, Dakota	1981		9				161		84						[16]	
	USA, Minnesota	1981		16				137		67						[16]	
	France	1982		40		5 17		370		40						[2]	
	France	1982						520								[2]	
	Germany	1979						63		22						[5]	
	Italy (1)	1979		4-15		74-111		37-74		333						[8]	

Table 3 : $^{210}\text{Pb}/^{226}\text{Ra}$ ratio in fly-ash from the emission control system of Megalopolis-IV Power Plant

Sampling Date	Economizer	LUVO	Rows of the left line of ESP				Rows of the right line of ESP			
			1 st	2 nd	3 rd	4 th	1 st	2 nd	3 rd	4 th
14-01-97 / 10:00	0.59	0.66	1.17	1.99	2.98	3.32	1.63	2.57	2.61	3.59
14-01-97 / 14:00	0.73	0.56	1.11	1.96	-	3.15	1.59	2.94	1.62	3.34
14-01-97 / 22:00	0.62	0.53	1.34	-	-	2.88	1.32	-	-	-
15-01-97 / 4:00	-	0.61	-	-	-	-	-	1.46	-	-
21-01-97 / 10:00	0.59	0.61	1.11	1.78	-	-	1.19	1.54	2.90	-
21-01-97 / 16:00	0.50	0.66	1.36	1.86	-	-	1.08	1.72	-	-
21-01-97 / 21:00	0.58	0.56	1.40	1.72	-	-	1.38	2.29	-	-
22-01-97 / 04:00	0.55	0.58	1.42	1.71	-	-	1.35	1.86	-	-
28-01-97 /10:00	0.66	0.58	1.07	2.14	2.24	-	1.32	1.62	2.03	-
28-01-97 /16:00	0.81	0.67	1.34	2.23	2.14	-	1.68	1.97	1.83	-
28-01-97 /21:00	0.73	0.57	1.58	1.94	2.10	-	1.48	1.92	2.02	-
29-01-97 /04:00	0.55	-	1.18	-	1.62	-	1.24	1.67	-	-
04-02-97 /10:00	-	-	0.78	-	2.03	-	0.96	1.94	2.02	-
04-02-97 /16:00	0.60	0.64	0.88	1.94	1.79	-	0.92	1.66	1.95	-
Mean value	0.63	0.60	1.2	1.9	2.1	3.1	1.3	1.9	2.1	3.5

Table 4 : $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{238}\text{U}/^{226}\text{Ra}$ activity ratio in fly-ash from Megalopolis-IV Power Plant

Sampling point		Sampling size	Radioactivity ratio	
			$^{210}\text{Pb}/^{226}\text{Ra}$	$^{238}\text{U}/^{226}\text{Ra}$
ECONOMIZER		15	0.60	0.89
LUVO		12	0.63	0.89
Left line	1 st row	27	1.2	0.98
	2 nd row	16	1.9	1.0
	3 rd row	13	2.2	1.1
	4 th row	4	3.1	1.3
Right line	1 st row	29	1.3	0.98
	2 nd row	22	1.9	1.0
	3 rd row	15	2.1	1.1
	4 th row	3	3.5	1.4

Table 5 : Field measurements in discarded fly-ash deposition fields in Megalopolis area.

Location	Description	Gamma-ray dose rate (nSvh ⁻¹)	²²² Rn exhalation rate (mBqs ⁻¹ m ⁻²)	²²² Rn in soil gas (kBqm ⁻³)	²²² Rn air concentration (Bqm ⁻³)
Deposition field 1	Restored in mid 70's, 40cm soil, planted.	150	18 ± 18	13 ± 5	26 ± 18
Deposition field 2	Restored in mid 80's, 40cm soil planted.	300	24 ± 50	42 ± 3	42 ± 24
Deposition field 3	Not yet restored, active (exhausted lignite mine)	450	2 ± 0.1	17 ± 3	19 ± 12
Nearby reference point	Undisturbed field	300	180 ± 120	600 ± 100	45 ± 28