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Radioenvironmental survey of the Megalopolis lignite field basin

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Abstract

The Megalopolis lignite field basin in southern Greece, with Megalopolis-A and B lignite-fired power plants in operation (total 900 MW), has been repeatedly investigated during the past 25 years by the Nuclear Engineering Section of the National Technical University of Athens (NES-NTUA). The present work aims at an integrated radioenvironmental approach leading to the dose assessment to the public and to the plants staff. This approach includes systematic sampling of lignite and barren at the local lignite mines feeding the power plants and sampling of lignite, fly-ash and bottom ash at the power plants for the determination of the activity of the natural radionuclides ²²⁶Ra, ²³²Th, ⁴⁰K, ²³⁴Th and ²¹⁰Pb. Furthermore, the following measurements and samplings were conducted in 25 selected sites within 10 km around the power plants: soil sampling for the determination of the above radionuclides, radon concentration and exhalation rate measurements, soil gas radon concentration measurements, dose measurements and calculations, determination of air-particulate matter concentration, etc. The results obtained allowed for the mapping of the parameters studied which lead to useful conclusions. Dosimetric calculations for the population living around the power plants staff were also performed based on the guidance of UNSCEAR (1982 report). © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Lignite-burning power plants; Radioenvironmental survey; Radon; Fly-ash

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1. Introduction

The Megalopolis lignite field basin, one of the two major lignite deposits under exploitation in Greece is located in the centre of the Peloponnese peninsula in southern Greece, near the city of Megalopolis (Fig. 1). Two lignite-burning power plants are in operation in this region: Megalopolis-A (600 MW - 3 units) since the early 70s and Megalopolis-B (300 MW - 1 unit) since the early 90s. The Megalopolis lignite is of very low calorific value and has rather high water and ash contents — it is amongst the poorest burned for electricity generation - resulting in relatively high fuel consumption per unit energy produced. Indicative consumption for Megalopolis-A power plant is approximately 2.3 kg of natural lignite per kWh produced (Simopoulos and Angelopoulos, 1987a). Moreover, Megalopolis lignite has higher natural radioactivity.

During the past 25 years, the natural radioactivity at the Megalopolis power plants has been extensively and repeatedly investigated throughout the whole fuel cycle, using highresolution/high-efficiency γ -spectroscopic setups, with Ge detectors by (Simopoulos and Angelopoulos, 1987a,b). During these investigations $^{226}Ra, ~^{232}Th$ and ^{40}K activity has been determined in the:

- 1. lignite and barren from the mines feeding the power-plants;
- 2. fly-ash and bottom-ash collected at the electrostatic precipitators (ESP); and
- 3. fly-ash collected from the stack.

Furthermore, in the frame of the natural radioactivity mapping of Greek surface soil, another wide research project of the Laboratory, 109 surface soil samples were collected within a 30 km distance from the power plants, allowing for the natural radioactivity mapping of the wider Megalopolis area (Anagnostakis et al., 1996), followed by dosimetric calculations from terrestrial gamma radiation based on UNSCEAR (1982). According to UNSCEAR the main pathways of radiation exposure of the plants staff and the population living around the plants from the radionuclides emitted from the stack, result from:

- 1. inhalation during the passage of the plume;
- 2. external exposure; and



Fig. 1. The principal population centres in the Megalopolis lignite field basin.

3. inhalation and ingestion resulting from the radionuclides deposited on the ground.

According to the same report the production of 1 GWa of electrical energy produced with coal burning leads to a total collective effective dose equivalent commitment of 2 manSv. This suggestion is based upon assumptions and estimations which take into account:

- 1. measured concentrations of natural radionuclides in coal and ash. The assumptions are made that the average activity concentrations in coal are 50 Bq kg⁻¹ of ⁴⁰K and 20 Bq kg⁻¹ for each of ²³⁸U and ²³²Th and that each of ²³⁸U and ²³²Th is in radioactive equilibrium with their precursors;
- 2. reported values of the efficiencies of the emission control system of the power plant; and
- 3. the population density around the plant.

Following the investigations conducted in the Megalopolis Power Plants (MPP), the following questions regarding the justification of the above UNSCEAR radiological assessment in the vicinity of lignite burning power plants were raised:

- natural radionuclide concentrations measured in lignite and the produced ashes may not be considered as representative of the whole life of a plant, but they are dependent upon the lignite deposit feeding the plant;
- 2. the efficiency of the emission control system of the power plants does not remain constant with time;
- 3. the dose exposure is not only due to the fuel cycle but also to the external γ -irradiation from the ground and radon exhalation from the ground, because of the increased natural radiation environment around a lignite deposit; and
- 4. fly-ash deposits also have a radioenvironmental impact.

Due to the above raised questions, a new integrated approach was undertaken to determine the significance of the above exposure pathways for better dose assessment to the public and to the plants staff.

2. An integrated radioenvironmental survey around a lignite burning power plant

In the frame of this research an integrated approach to the radioenvironmental survey of the Megalopolis lignite field basin was planned and conducted. The survey which may be applied in the case of any coal/lignite burning power plant, consists of the following steps:

- 1. Systematic sampling for a certain period of time, of:
 - 1.1. lignite and barren at the mine(s) feeding the power plants; and
 - 1.2. lignite and ash from the power plants under investigation, to allow for the determination of representative values for the natural radioactivity content of the above materials.
- 2. Investigation of the input (lignite) and output (ashes) of the plants, in order to investigate their radioactivity balance through the combustion process which will subsequently lead to the estimation of the radioactivity releases to the environment.
- 3. Grid survey around the power plants. In each sampling location the following samplings and measurements were conducted:
 - 3.1. sampling of the surface and 0–80 cm soil layer to allow for the mapping of the surface soil activity of the natural radionuclides (²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th and ⁴⁰K), study of their vertical distribution and study of the existence of radioactive equilibrium among the nuclides of the natural radioactive chains;
 - 3.2. soil gas radon concentration measurements, together with soil permeability measurements;
 - 3.3. radon exhalation measurements from the surface of the ground;
 - 3.4. ambient air radon concentration measurements;

- 3.5. particulate matter air concentration; and
- 3.6. in-situ gamma-ray dose rate measurements.

Monitoring of meteorological conditions in each sampling location during the measurement and sampling time is essential for the subsequent validation and correlations of the results obtained. Such an approach may lead to:

- 1. mapping of the fly-ash deposition patterns within a critical area around the plants;
- 2. dose assessment calculations for the plants staff and the population living around them, due to fly-ash release;
- 3. dose assessment calculations due to other factors which are not directly related to the power plants; and
- 4. modelling of the radon transport from the ground to the atmosphere, in the vicinity of lignite deposits with increased natural radioactivity.

In the frame of the proposed survey the following samplings and measurements have been conducted in the present work:

- 1. The collection of 40 lignite and barren samples from the local open lignite mines feeding the power plants. All samples were analysed using high resolution/high-efficiency γ -spectroscopic set-ups with Ge detectors, for the determination of ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th and ⁴⁰K activity; details on the γ -spectroscopic analyses conducted may be found in (Simopoulos and Angelopoulos, 1987a).
- 2. Lignite feeding the Megalopolis-B power plant and fly and bottom ashes produced in the same power plant. A total of 6 weekly representative samples of each material were prepared covering a consecutive 6-week period. Each weekly sample was prepared from daily collected samples as described in (Simopoulos and Angelopoulos, 1987a).
- 3. A total of 350 fly-ash samples were collected at 18 different stages such as the emission control system, the electrostatic precipitators

(ESP), the economizer, etc., of the Megalopolis-B power plant. Almost half of the samples have already been analysed for the determination of 226 Ra, 232 Th and 40 K activity using high-resolution/high efficiency Ge detector set-ups. For many of the samples which were analysed with a LEGe detector, 238 U and 210 Pb activity was also determined.

- 4. Grid sampling within 10 km around the power plants. A total of 25 sampling locations were selected as shown in Fig. 1. In every sampling location the following samplings and measurements were conducted:
 - 4.1. Surface soil sampling (1 cm depth) and soil samples from 0–20, 20–40, 40–60 and 60–80 cm depth for the study of the vertical profile of the radionuclides under study. The samples were analysed for the determination of 238 U, 226 Ra, 210 Pb, 232 Th and 40 K activity.
 - 4.2. Soil gas radon concentration measurements, at a depth of 80 cm. Soil permeability measurements.
 - 4.3. Radon exhalation from the surface of the ground.
 - 4.4. Ambient air radon concentration measurements.
 - 4.5. Particulate matter concentration in air. Two high volume air-samplers were simultaneously used in each location in order to determine the particulate matter in air 1.6 m above the ground. Gamma spectroscopic analysis of the collected filters followed to determine any detectable natural radionuclides, such as ²¹⁰Pb. The filters will also be analysed using XRF and NAA for the determination of heavy metals.
 - 4.6. Gamma-ray dose rate measurements 1 m above the ground, using a portable NaI detector, calibrated for dose (in $nSv h^{-1}$).
 - 4.7. In-situ gamma-spectroscopic measurements using a stabilized $3'' \times 3''$ NaI detector system 1 m above the ground. In each location a spectrum was collected for at least 1 h. The analysis of the spectrum, together with the dose

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measurements and the results of the gamma-spectroscopic analysis of the soil samples collected lead to very useful results regarding the exposure due to terrestrial radiation and presumably due to radon daughters concentration in the air.

- 4.8. Monitoring of meteorological conditions such as: ambient temperature and moisture in ground level, and wind speed and direction at 5 m height from the ground, using a portable meteorological station on-line connected to a computer. A computer program was written and used to provide integrated results of wind speed and direction over a period of 10 min.
- 5. Analysis of spring water for the determination of radon concentration. Samples were analysed by a portable measuring system for the direct determination of the radon concentration in water.

3. Results

The results of the gamma spectroscopic analysis of the lignite and barren from the mines, and the lignite feeding the power plant Megalopolis-B and the ashes produced are presented in Table 1. It is important to mention that significant disequilibrium among ²¹⁰Pb and ²²⁶Ra was observed in the case of certain fly ash samples, which were found highly enriched in ²¹⁰Pb, with the ratio ²¹⁰Pb/²²⁶Ra reaching the value of 4, depending on the sampling location along the emission control system of the Megalopolis-B power plant. This is attributed to the different physico-chemical properties of ²¹⁰Pb and ²²⁶Ra, and their different behaviour during the combustion process. This disequilibrium should be taken into account when sampling inside a lignite burning power plant and when performing dosimetric calculations for the plant's staff. Furthermore, according to UNSCEAR (1982) this disequilibrium is expected to be higher in the fly-ash escaping the power plant.

It is important to note that ²²⁶Ra, ²³²Th and ⁴⁰K activity of lignite feeding the Megalopolis-B power plant and the fly-ash produced estimated in the frame of this research compared well with the values obtained 12 years ago (Simopoulos and Angelopoulos, 1987a) for Megalopolis-A. They show that there is no significant difference in the natural radioactivity between the lignite feeding the two plants and the ashes produced.

Table 2 presents the results of the measurements performed in the 25 locations around the power plants. For comparison purposes, the results obtained from similar measurements performed in two reference locations are presented, namely:

- 1. Athens, Zografos, The National Technical University of Athens Campus; and
- 2. The Lavrion lead mine (now exhausted), 50 km south-east of Athens.

The amount of available data allowed for the mapping of the measured parameters. The in-

Table 1

Gamma spectroscopic analysis results of lignite, fly-ash and bottom-ash from Megalopolis-B power plant (mean values for a 6-week sampling period)

Material	Mean activity over a 6-week sampling period $(Bq kg^{-1}) \pm total error (\%)$						
	²³⁸ U (²³⁴ Th)	²²⁶ Ra	²¹⁰ Pb	²³² Th (²²⁸ Th)	⁴⁰ K		
Lignite	306 ± 13	346 ± 8	361 ± 10	19 ± 9	173 ± 14		
Fly-ash	964 ± 7	904 ± 9	1158 ± 11	52 ± 2	454 ± 11		
Bottom ash	681 ± 4	662 ± 9	275 ± 6	41 ± 5	405 ± 11		

Table 2

	Units	Range		Reference data range	
		0-5 km	5-10 km	Value	Site ^a
Surface soil ²²⁶ Ra concentration	$(Bq kg^{-1})$	26-337	23-42	9-20	1
				18-53	2
Surface soil ²³² Th concentration	$(\mathrm{Bq} \mathrm{kg}^{-1})$	24-41	12-43	10-23	1
				7-38	2
Surface soil 40 K concentration	$(Bq kg^{-1})$	154-477	218-631	168-238	1
				39-485	2
External γ -ray dose rate of	$(nSv h^{-1})$	40-187	28-72	17-33	1
terrestrial origin, due to: 40 K, 232 Th, 226 Ra, (calculated)				14-68	2
External γ-ray dose rate (measured)	$(nSv h^{-1})$	63-331	50-180	45-60	1
Soil gas radon	$(kBq m^{-3})$	4-90	2-81	0.2 - 2.8	1
(monitored at 0.6-0.8 m)				0.3-1.6	2
Soil permeability	(m^2)	$10^{-15} - 10^{-9}$	$10^{-14} - 10^{-9}$	$10^{-12} - 10^{-11}$	1
1	. ,			$10^{-13} - 10^{-11}$	2
Surface soil exhalation rate	$(mBq m^{-2}s^{-1})$	0 - 2166	0-230	0-12	1
				7-122	2
Radon in air concentration	$(Bq m^{-3})$	0-835	0-222	15-102	1
				0-200	2
Particulate matter air-	$(\mu g m^{-3})$	25-248	18-124	139	1
concentration	N 0 /				2
Radon in water concentration	$(Bq l^{-1})$		5-26	0-10	1

Results of the survey within the 10 km radius from the Megalopolis power plants (25 sampling locations)

^aSite 1: Athens, Zografos, The National Technical University of Athens Campus. Site 2: Lavrion Lead Mine (now exhausted), 50 km south-east of Athens.

house built complex Data Base/Geographical Information System (DBGIS) was used, to produce the following mappings:

- 1. ²²⁶Ra, ²³²Th and ⁴⁰K activity of the 1 cm surface soil, (Figs. 2–4, respectively).
- 2. External gamma-ray dose rate of terrestrial origin due to ²²⁶Ra, ²³²Th and ⁴⁰K (Fig. 5), calculated according to UNSCEAR (1993) suggestions from the formula:

$$D = 0.0414 \cdot A_{\text{K}-40} + 0.623 \cdot A_{\text{Th}-232} + 0.461 \cdot A_{\text{Ra}-226}$$

where *D* is the absorbed dose rate in air in nGy h^{-1} and A_{K-40} , A_{Th-232} and A_{Ra-226} the activity concentrations in soil of 40 K, 232 Th and 226 Ra respectively, in Bq kg⁻¹.

- 3. External gamma-ray dose rate measured on the site (Fig. 6).
- 4. Radon concentration in the soil gas, radon

exhalation rate from the soil and radon concentration outdoors (Figs. 7–9).

Given the ²²⁶Ra content of the lignite, the ash content, the partitioning between slag and fly ash and the efficiency of the emission control systems, the ²²⁶Ra radioactivity discharges from Megalopolis-B power plant are estimated to be approximately 3500 MBq (Gwa)⁻¹, which is much lower than those of the Megalopolis-A plant, which is approximately 52 000 MBq (Gwa)⁻¹ (Simopoulos and Angelopoulos, 1987a); this is attributed to the significant difference in the efficiency of the electrostatic precipitation devices between the two plants.

4. Discussion and conclusions

The following conclusions may be drawn from the measurements and analysis of samples:



Fig. 2. Surface soil (1 cm) ²²⁶Ra activity mapping of the Megalopolis lignite field basin.



Fig. 3. Surface soil (1 cm) ²³²Th activity mapping of the Megalopolis lignite field basin.



Fig. 4. Surface soil (1 cm) ⁴⁰K activity mapping of the Megalopolis lignite field basin.



Fig. 5. External gamma-ray dose rate of terrestrial origin due to 226 Ra, 232 Th and 40 K in the Megalopolis lignite field basin, calculated according to UNSCEAR (1993).



Fig. 6. External gamma-ray dose rate measured in the Megalopolis lignite field basin.

- 1. Within 10 km radius from the plants there is a hot-spot of ²²⁶Ra activity on surface soil, which is attributed to the fly-ash deposition. A similar increase in ²³²Th and ⁴⁰K activity was not observed.
- The calculated and the measured gamma-ray 2. dose rate, although they both indicate the same hot spot, have significantly different values within the 10 km area from the power plants, with the measured dose sometimes being two times higher than the calculated. Outside the 10 km area the calculated and measured doses agree satisfactorily. This is an indication that the dose conversion factors suggested by UNSCEAR (1993) may not be valid for dose estimations in the vicinity of a lignite mine or a lignite power plant. The calculated dose is relatively high compared with the dose estimated for the whole of Greece (Fig. 7).
- 3. Radon concentration in soil gas is increased

inside the 10 km radius compared with that at the two reference sites, with a hot spot located in the vicinity of one of the open lignite mines under exploitation. Radon concentration outdoors (Fig. 10) has increased values within the 5 km radius compared with the two reference sites, with a pattern similar to that of soil gas radon concentration.

4. Radon exhalation rate from the ground surface within the 10 km was increased compared with that at the two reference sites. The respective pattern does not agree well with those of soil gas radon concentration and radon concentration outdoors, but agrees with the pattern of ²²⁶Ra concentration on surface soil. Possible reasons for this disagreement are: hill effects in the southern part of Megalopolis region, meteorological conditions and the existence of fly-ash deposits. However, further investigation is needed.



Fig. 7. External gamma-ray dose rate of terrestrial origin in Greece due to 226 Ra, 232 Th and 40 K, calculated according to UNSCEAR (1993).



Fig. 8. Radon concentration in the soil gas in the Megalopolis lignite field basin.



Fig. 9. Radon exhalation rate from the soil in the Megalopolis lignite field basin.



Fig. 10. Radon concentration outdoors in the Megalopolis lignite field basin.

5. Particulate matter air concentration was increased within the 5 km radius, while within the 5-10 km radius it was not significantly different compared with that at the two reference sites.

5. Dosimetric calculations

Based on the above measurements some dosimetric calculations were applied. Using the recorded external gamma-ray dose rate 125-250 nSv h^{-1} and the total population of 12000 people living within 10 km, the resulting collective dose ranges between 13 to 26 manSv year⁻¹. The collective dose for the whole Greek population (\sim 11×10^6 people), for a mean gamma-ray dose rate 46 nSv h^{-1} is estimated to be 4400 manSv year⁻¹. According to UNSCEAR (1982) estimations, the Collective Effective Dose Equivalent Commitment from external irradiation, due to activity deposited following the power plants atmospheric releases for the total power generated in the Megalopolis region is $0.081 \text{ manSv year}^{-1}$, which is very low compared with the collective dose calculated from published doses.

Assuming the worst case scenario, that all air particulates detected on the most heavily contaminated air filter, collected close to an open lignite mine are fly-ash particles with the highest 226 Ra activity 1000 Bq kg⁻¹, the resulting 226 Ra activity concentration in air does not exceed the level of 250 mBq m⁻³. This value is high compared with similar measured values 50 μ Bq⁻³ or normal values 1 μ Bq m³ (UNSCEAR, 1982), but is far below the Derived Air Concentration Limit of 10 Bq m⁻³ suggested by the ICRP (1978) for occupational exposure.

Taking into account the highest value of the

equilibrium equivalent radon concentration of 500 Bq m⁻³ recorded outdoors in the Megalopolis region, dosimetric calculations performed according to UNSCEAR (1982) resulted in an effective dose equivalent commitment of 15.5 mSv year⁻¹ which is high compared with the estimated dose of 0.015 mSv due to radon and radon daughters from power plants atmospheric releases, calculated according to UNSCEAR (1982), for the Megalopolis region.

Future work should be focused on the investigation and the justification of the radiological assessments, suggested by UNSCEAR (1982). The dosimetric modelling of the plants staff and the population living around the power plants will be further investigated taking into consideration all the collected data. Furthermore, the various radioenvironmental processes within the lignite basin will be modelled by extending and tuning existing appropriate numerical models.

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