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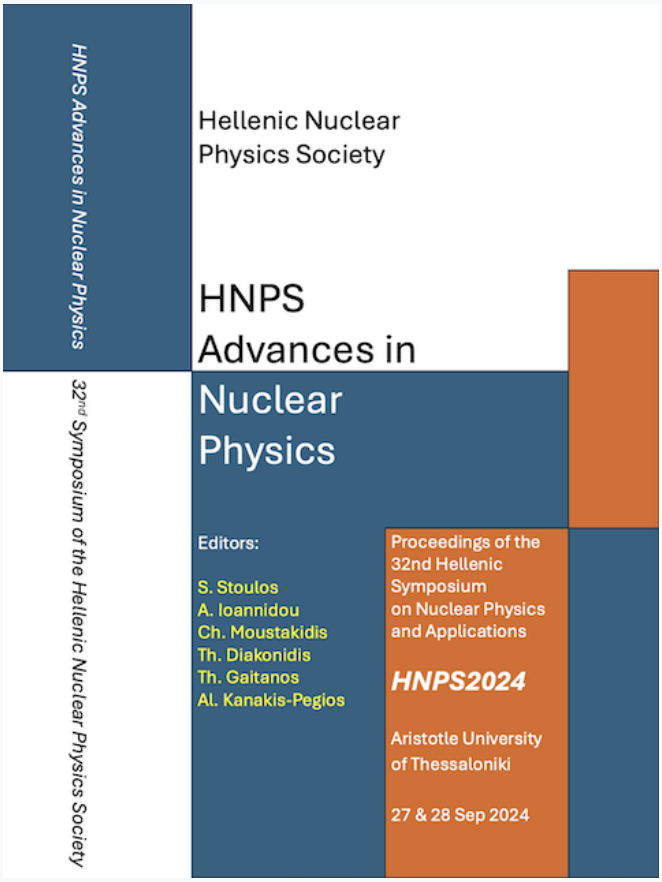

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Comparative Measurements of Greek Lignite Ash and Disposal Materials

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Abstract Coal power plants produce big volumes of ash via the combustion procedure. Most of the fly ash is detained with appropriate filters and the bottom ash gathers at the lower levels of the combustion chamber. Large volumes of ash are disposed to refill the old coal mines, and a small part is exploited in the cement production industry. The refilling material mostly contains fly ash and material which consists of soil and negligible amounts of coal and therefore not useful for power production.

This study compares the concentrations of radionuclides in fly ash samples before and after the deposition as a refilling material. The method of measurement involves preparation of the sample and γ spectroscopy with High Purity Germanium detector. Higher concentrations in pure ash appeared for the radionuclides ^{226}Ra and ^{40}K, 318 Bq/kg and 382 Bq/kg, respectively. In addition, comparison among the different regions of ash deposition is conducted. Even samples from the same area differ in radionuclides' concentration. The results are in line with older studies, ordered by Public Power Corporation, and reveal that the concentrations are close to or below the clearance levels, set by the Euratom directive of 2013.

Keywords Naturally Occurring Radioactive Material (NORM), Lignite Combustion, Environmental Radioactivity, Climate Change, Coal Power Plant

INTRODUCTION

For over seven decades, lignite has been a fundamental pillar of Greece's electricity production. Since the 1950s, this abundant, low-grade coal has powered industries, homes, and infrastructure, serving as a readily available and cost-effective energy source. The country's richest lignite deposits are concentrated in Western Macedonia—particularly in Ptolemaida, Amyntaio, and Meliti—as well as in the Peloponnese, around the Megalopolis region. These areas have been at the heart of Greece's energy landscape, sustaining power generation and employment opportunities for decades.

However, lignite combustion presents significant environmental challenges, primarily due to the production of ash. For every ton of lignite burned, between 100 to 300 kg of ash are generated and specifically for the lignite extracted in Greek mines, 16% of its mass turn into ash [1]. Lignite ash is a Naturally Occurring Radioactive Material (NORM) and can be categorized into two types: bottom-ash, which settles at the base of the combustion chamber, and fly-ash, which is captured by filtration systems before being released into the atmosphere. The presence of ash, along with potential variations in lignite quality, affects the efficiency of power plants and contributes to environmental issues such as air pollution and land degradation.

As Greece transitions towards cleaner and more sustainable energy sources, in the last two decades the role of lignite is gradually being reconsidered. With increasing efforts to reduce carbon emissions and invest in renewable energy, lignite's dominance in the country's energy mix is gradually declining. However, its historical significance and the challenges associated with its use continue to raise discussions on energy policy, environmental protection, and economic transformation.

Over the years, using lignite to generate electricity in Greece has led to a total production of over hundreds million tons of ash. A small portion of this ash has been recycled in the cement-production industry, while the majority of it has been damped along with waste soil material in the old, depleted

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lignite mines. Therefore, the damping sites are in the same areas: in Western Macedonia (Ptolemaida, Amyntaio, Meliti) and in Peloponnese (Megalopolis). In the past the concentrations of natural radioactivity of Greek ash and lignite were investigated thoroughly by Simopoulos, Angelopoulos [2], Manolopoulou, Papastefanou [3], Skordas et al [4], Tsikritzis et al. [5]. The present study contributes to the series of these works, as the most recent one.

MATERIALS AND METHODS

In this study 20 samples were collected from the aforementioned areas in the period from October 2023 to May 2024. The samples consist of three different material categories:

- Lignite (the raw material that was used for combustion and power production)
- Ash (bottom ash and fly ash)
- Disposal material, which is the refilling material of the depleted lignite mines. This material contains fly ash along with useless material which extracted during lignite mining but contained negligible amounts of coal and therefore was not useful for combustion.

Lignite sample originated from the mines of Ptolemaida. The ash samples were: (a) two samples from the ash deposition at Mavropigi depleted mine, which is near Ptolemaida, and two samples from the ash deposition at Megalopolis depleted mine, (b) two samples directly from the ash production of Ptolemaida-5 power plant (i.e. one bottom-ash sample and one fly-ash sample). Eleven disposal materials' samples were collected from three different depleted mines at Ptolemaida (seven samples), Amyntaio (one sample) and Meliti (three samples).

The samples were measured with High Purity Germanium (HPGe) γ-spectroscopy with 50% relative efficiency, to determine the radioisotopic concentration of natural (^{238}U, ^{232}Th, ^{226}Ra, ^{40}K) and anthropogenic (^{137}Cs) radioactivity. The samples were air dried and smashed into powder. The measurement was carried out for approximately 12-24 hours.

For the determination of the activity concentration of ^{137}Cs, the energy photopeak of 662 keV was used, while for ^{40}K, the unique energy photopeak of 1461 keV was employed. Considering secular equilibrium and that the equilibrium of the natural radioactivity series is not disturbed, (a) ^{232}Th was determined by calculating the concentration of ^{228}Ac, utilizing the 911 keV photopeak, (b) ^{238}U was determined by its daughters: ^{234}Th emitting γ-rays at 63 keV, and ^{234}Pa emitting γ-rays at 1001 keV. The concentration of ^{226}Ra was calculated as the median of the calculated concentrations derived by the following methods: (a) Subtraction of the ^{235}U-counts by the 186 keV photopeak. These counts were calculated by the constant percentage of ^{235}U in natural uranium and the measured photopeaks of 63 keV and 1001 keV, ultimately corresponding to ^{238}U. (b) Gilmore correction (Gilmore 2008), considering 57.1% of the counts at 186 keV originates from ^{226}Ra.

The concentration of activity (Bq/kg) was derived by the formula:

$$A(Bq/kg) = \frac{cps}{m * I\gamma * eff} \quad (1)$$

Where cps is the net counts per second captured under each photopeak, m is the mass of the sample, Iγ is the probability of the γ-emission by the nucleus and eff is the efficiency of the HPGe for the specific γ-photon energy. The propagation of the uncertainties of these quantities to the derived concentration A, was calculated by the formula:

$$\sigma A = A(Bq/kg) * \sqrt{\left(\frac{\sigma m}{\mathrm{m}}\right)^2 + \left(\frac{\sigma I\gamma}{\mathrm{I\gamma}}\right)^2 + \left(\frac{\sigma cps}{\mathrm{cps}}\right)^2 + \left(\frac{\sigma eff}{\mathrm{eff}}\right)^2} \quad (2)$$

Where σm, σcps, σIγ, σeff are the standard deviation of mass, cps, Iγ and eff, respectively.

RESULTS AND DISCUSSION

The lignite radioisotopic composition can be the base of comparison of the three different material categories, as it originates from the local ores. Figure 1 illustrates the concentration of the radionuclides in the lignite before it undergoes combustion in Ptolemaida 5 coal power plant. Lignite sample exhibits low levels of natural radioactivity with concentration values of ^{238}U at 87 Bq/kg, ^{232}Th at 12 Bq/kg, ^{226}Ra at 36 Bq/kg, ^{40}K at 127 Bq/kg. These radioisotopes occur naturally in the earth's crust.

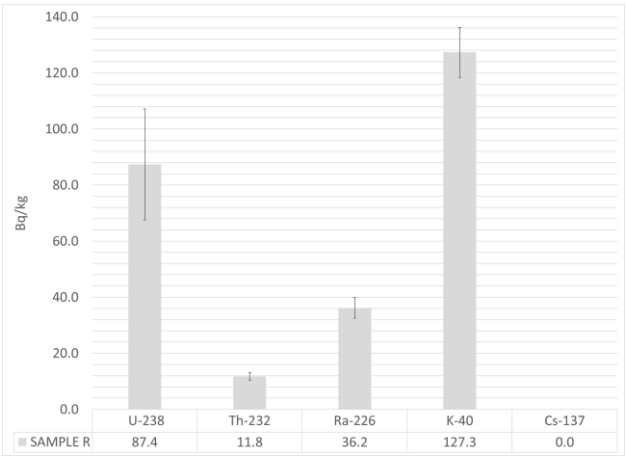


Figure 1. *Sample R is the Lignite sample from Ptolemaida 5 Power Plant.*

The concentration of anthropogenic radionuclide ^{137}Cs was found to be negligible, as it did not penetrate the very first layers of the ground where it was deposited from the era of the Chernobyl accident and thus cannot reach the depths where the lignite ores occurred [6].

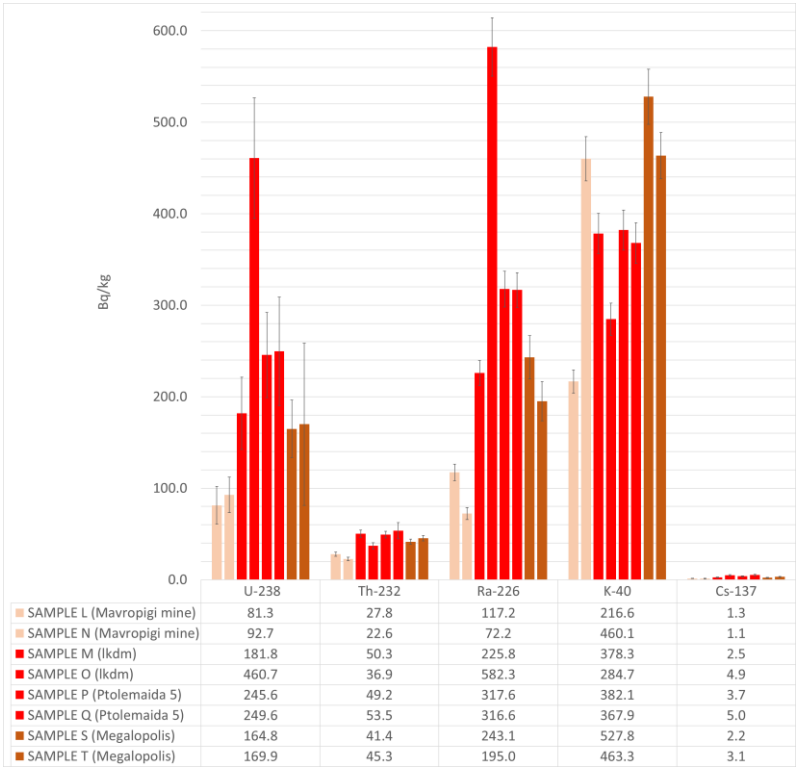
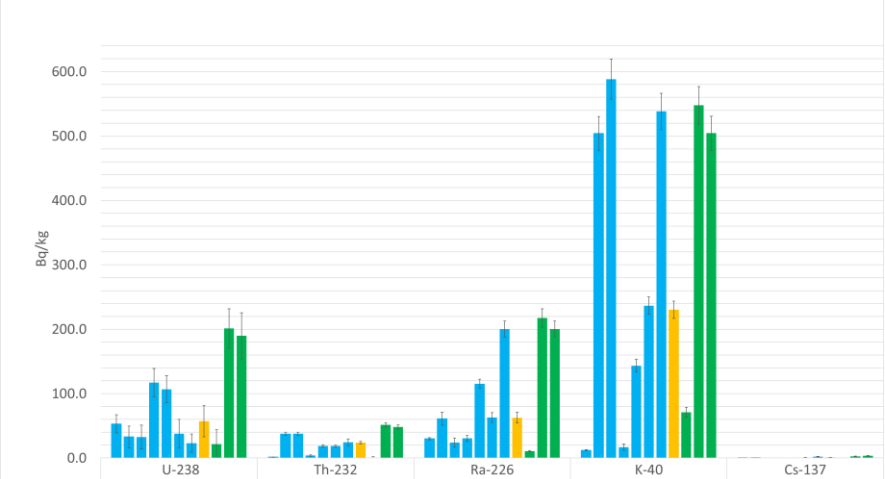


Figure 2. *Ashes collected from the depositions in the depleted mines and directly from Ptolemaida-5*

The ash samples which were collected from old depositions of ash in depleted mines (samples L, N, S, T in Fig. 2) exhibit a higher concentration of ^{40}K. The higher concentration of U series isotopes in Megalopolis deposits is because of the much higher concentrations of U series isotopes in the

Megalopolis lignite [3,4]. Ash samples collected from the combustion chamber and samples M, O, which were collected from deposition of "fresh" ash in depleted mine, show higher concentrations of ^{226}Ra, ^{238}U and ^{232}Th than of ^{40}K.

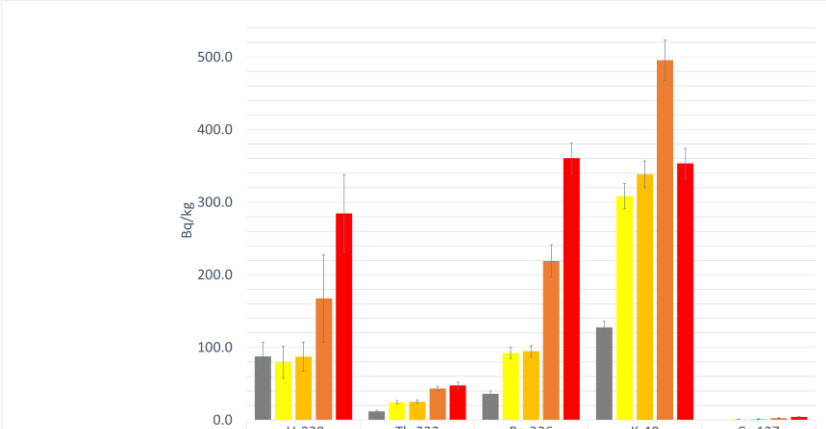
The disposal materials (Fig. 3), which were collected from depleted mines have lower concentrations than the ash samples, probably due to the, not for radiological purposes, mixing of the ash with useless material which had been extracted along with lignite from the old mines. Once again, ^{40}K is in higher concentrations in the old mines' material, than in the "fresh" ash.



	U-238	Th-232	Ra-226	K-40	Cs-137
SAMPLE A	53.1	1.5	30.0	12.1	0.1
SAMPLE B	32.8	37.5	61.1	504.2	0.1
SAMPLE C	32.5	37.6	24.0	588.3	0.0
SAMPLE D	117.2	3.8	30.4	16.9	0.0
SAMPLE E	106.9	18.6	115.4	143.6	0.0
SAMPLE F	37.9	18.6	63.1	236.6	0.9
SAMPLE G	23.1	24.5	200.5	538.3	2.2
SAMPLE H	57.3	23.9	62.7	230.3	1.1
SAMPLE I	21.8	0.4	10.8	71.0	0.1
SAMPLE J	201.1	51.5	217.3	547.7	2.7
SAMPLE K	189.7	48.2	200.5	504.5	3.3

Figure 3. *Disposal material samples from the depleted mines of Ptolemaida (blue), Amyntaio(orange) and Meliti (green)*

In Figure 4, a comparison among the samples of (a) lignite, (b) disposal materials, (c) ash from Macedonia deposition, (d) ash from Megalopolis deposition, and (e) ash from combustion chamber is presented. Where two or more samples were available the average value is presented.



	U-238	Th-232	Ra-226	K-40	Cs-137
LIGNITE(WESTERN MACEDONIA)	87.4	11.8	36.2	127.3	0.0
DISPOSAL MATERIALS(WESTERN MACEDONIA)	79.4	24.2	92.3	308.5	1.0
ASH FROM DEPOSITION(WESTERN MACEDONIA)	87.0	25.2	94.7	338.4	1.2
ASH FROM PELOPONNESE	167.4	43.3	219.1	495.6	2.6
ASH(WESTERN MACEDONIA)	284.4	47.5	360.6	353.3	4.0

Figure 4. *Comparison of concentrations of lignite, ash and disposal material*

To reveal the transfer of the radionuclides from lignite to ash, an assumption, that the ash retains all the radionuclides of the lignite, can be made initially. Therefore, the natural radioisotopic transferring from lignite to ash can be calculated by: (a) assuming that 16% of the mass of the dry lignite transforms into ash after the combustion in the "Ptolemaida 5" Coal Power Plant and (b) the natural radioactivity concentrations (Bq/kg) of lignite, which is presented in Figs. 1 and 4. In other words, the whole activity of 1 kg of lignite is captivated in 0.16 kg of ash. These calculated concentrations of natural radioactivity in the produced ash are presented in Fig. 5 with grey color. Comparing them with the concentrations of the real samples of ash (red color) and considering the coefficient of variance of the real samples, can derive useful conclusions. For instance, after the combustion of lignite: ^{40}K, ^{238}U and ^{232}Th do not remain fully in the produced ash, while ^{226}Ra remains fully. Detailed mapping of concentrations at the surrounding area and γ-spectroscopy of additional samples from the combustion chambers may reveal the escaping-from-the-ash path. A preliminary conclusion can be extracted by the comparison of ^{40}K in the depleted mines' matter (Figures 2 and 3) with ^{40}K in the bottom ash. Further investigation of the above assumption and calculations may include the findings of relevant works performed in the past [2-5], including comparison of the calculated concentration with a plethora of measured ones.

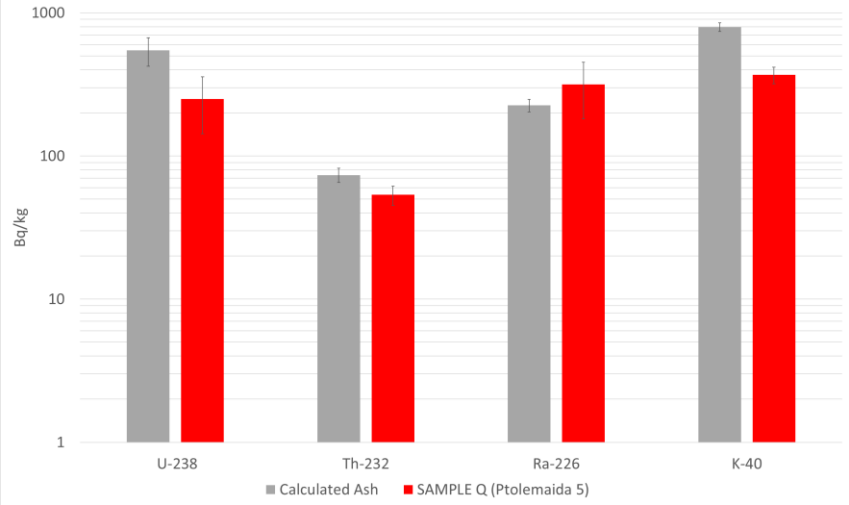


Figure 5. *Comparison of theoretically calculated concentrations with measured concentrations in bottom ash (sample Q) for the "Ptolemaida 5" CPP. Propagation of uncertainty and coefficient of variance is used for the error-bars of calculated and measured concentration, respectively.*

CONCLUSIONS

The concentrations of natural radioactivity in the disposal materials and in the ash samples are well below the limits of the EURATOM Directive-2013/59 (see Table A.2 of ANNEX VII) [7]. The management of the ash deposition has been safely conducted through the years, to reduce any consequences in public health. Greek lignite has low concentrations of radioactivity and thus the activities of the produced ash are well below the global average.

Most of the radioactivity of the original lignite ore remains in the produced ash after the combustion. Further studies may confirm the initial conclusion that a small part of the radionuclides ^{40}K, ^{238}U and ^{232}Th do not remain fully in the produced ash but maybe escape in the environment.

Acknowledgments

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