

Radiation Dose Rates and Exposure Associated to Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) at Locations of a Portuguese Coal-Fired Power Plant – 14431

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ABSTRACT

Coal contains trace quantities of natural radionuclides such as Th-232, U-235, U-238, as well as their radioactive decay products and ^{40}K . These radionuclides can be released as fly ash in atmospheric emissions from coal-fired power plants, dispersed into the environment and deposited on the surrounding top soils. Therefore, the natural radiation background level is enhanced and consequently increase the total dose for the nearby population.

A radiation monitoring programme was used to assess the external dose contribution to the natural radiation background, potentially resulting from the dispersion of coal ash in past atmospheric emissions. Radiation measurements were carried out by gamma spectrometry in the vicinity of a Portuguese coal-fired power plant. The radiation monitoring was achieved both on and off site, being the boundary delimited by a 20 km circle centered in the stacks of the coal plant.

The measured radionuclides concentrations for the uranium and thorium series ranged from 7.7 to 41.3 Bq/kg for Ra-226 and from 4.7 to 71.6 Bq/kg for Th-232, while K-40 concentrations ranged from 62.3 to 795.1 Bq/kg. The highest values were registered near the power plant and at distances between 6 and 20 km from the stacks, mainly in the prevailing wind direction. The absorbed dose rates were calculated for each sampling location: 13.97-84.00 $\eta\text{Gy/h}$, while measurements from previous studies carried out in 1993 registered values in the range of 16.6-77.6 $\eta\text{Gy/h}$. The highest values were registered at locations in the prevailing wind direction (NW-SE).

This study has been primarily done to assess the radiation dose rates and exposure to the nearby population in the surroundings of a coal-fired power plant. The results suggest an enhancement or at least an influence in the background radiation due to the coal plant past activities.

INTRODUCTION

Radioactivity dispersed in the environment results not only from the nuclear fuel cycle activities

but also from other several technological activities. The most relevant non-nuclear industry, in what concerns to radioactive elements emissions, are phosphates industry, ceramic industry and energy production activities, in particular thermal coal power plants.

Coal contains trace quantities of naturally occurring radionuclides like K-40, Th-232, U-235 and U-238 as well as the radioactive decay products of the last three nuclides. Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) are generated when burning removes organic constituents, leaving minerals and concentrating trace quantities of these radionuclides and their radioactive decay products, including radium, in ash. The amount of Ra-226 in ash can vary by more than two orders of magnitude depending on the type of coal (composition in organic matter) and where it was mined [1]. Ash also typically contains silicon, aluminium, iron and calcium. These elements make up about 80 to 90% of all of the constituents of coal ash in addition to other trace elements: As, B, Cd, Cr, Ni, Pb, Se, Zn, Hg e V. These trace elements can be enriched between 2 and 100 times relative to the original coal concentration.

About 10 percent of the original volume of coal will result in ashes generated in the combustion process: fly ash (74%), bottom ash (20%) and boiler slag (6%) [1]. Fly and bottom ashes make up the majority of the coal combustion by-products and although they have the same origin they are physical and chemically different. Fly ash is the finest portion of coal ash particles, with diameter typically between 10 and 100 microns; it is transported from the combustion chamber by exhaust gases and removed by electrostatic precipitators, baghouses, and scrubbers to reduce the emissions to the atmosphere by at least 95 % [1]. Bottom ashes consist of larger (and therefore heavier) particles collected at the bottom of the furnace.

Coal-fired power plants equipped with sophisticated retention devices release about 1% of the total fly ash produced. However, huge amounts of coal are burnt every year (millions of tonnes) and therefore 1% can be a significant amount of fly ash released with enhanced radionuclides concentrations [1], [2]. Moreover, in the past, the boilers were not equipped with efficient control devices so fly ash retention efficiency was much lower; throughout the world, the proportion of fly ashes discharged into the atmosphere ranged from 0.5 to 20% in the past decades [2].

The activity concentrations of natural radionuclides which are released into the atmosphere from a coal power plant will depend on many factors, such as the activity in coal, ash content inert matter of the coal, temperature of combustion, portioning between bottom ash and fly ash and efficiency of the filtering system [3]. Therefore, marked differences should be expected between the by-products produced and the amount of activity discharged, per unit of energy produced, from different coal fired power plants.

People living near coal power plants may be exposed to increasing quantities of radioactive isotopes through inhalation, external irradiation and ingestion. As the half-lives of these radionuclides are practically infinite in terms of human lifetimes, the accumulation of these

species in the biosphere is directly proportional to the length of time that a quantity of coal is burned [1]. Several authors have studied the possible influence on the level of natural radioactivity in the surrounding environment of coal-fired power plant [4], [5], [6], [7], [8], [9], [10], [11], [12], [13]. The results from these studies are not unanimous.

This work describes the methodology developed to assess the radiation dose rates and exposure associated to the technologically enhanced naturally occurring radioactive materials (TENORM) from a Portuguese coal-fired power plant. A radiation monitoring programme was used to assess the external dose contribution to the natural radiation background, potentially resulting from the dispersion of coal ash in past atmospheric emissions.

MATERIALS AND METHODS

The study area

The present research was carried out in the surroundings of a coal-fired power plant located along the south-western coast of Portugal. The coal plant is located near a dense populated area, at about 6 km to southeast from the city of Sines and about 150 km south of the capital, Lisbon (Fig. 1). It has been operational since 1980s, fueled by bituminous coal in four coal fired units with a capacity of 314 MWe each one; the first unit started working in 1985 and the last one in 1989 [1], [14]. The plant has two operational stacks, both with 225 m height.



Fig. 1: Sines Coal-Fired Power Plant Location.

This coal-fired power plant consumes, on average, 2.55 Gkg of coal per year. In 2011 it consumed 2.64 Gkg (0.36 Mkg/GWh) for an energy production of 7432 GWh, approximately 0.29 Mkg of particulate matter were released into the atmosphere, 39.77 Mkg of non-conformity fly ash was conducted to disposal into the coal plant landfill as well as 26.38 Mkg of bottom slag [15].

By the end of 2011, the amount of accumulated material (fly ash and bottom slag) in the plant disposal was 1.09 Gkg, covering an area of about 0.11 km². In 2009 and 2010 the coal consumption was about 3.19 and 1.82 Gkg, respectively. Coal is stored onsite in four active uncovered piles with 150 Mkg each and in one passive pile with 7 Gkg [1], [15].

The European database “European Pollutant Release and Transfer Register” contains data on this particular source emission for the years of 2001, 2004, 2007, 2008, 2009, 2010 and 2011 [16] (Table I). The data refers to PM₁₀ (Fig. 2) heavy metals and others pollutants releases into air.

TABLE I: Pollutant Releases to Air from Sines Coal-Fired Power Plant (kg) [16].

Releases to air (kg)	2001	2004	2007	2008	2009	2010	2011
Arsenic and compounds	48	727	40	31	33	-	27.2
Cadmium and compounds	12	-	-	-	-	-	-
Chlorine (as HCl)	738x10 ³	-	103x10 ³	21.8x10 ³	833x10 ³	17.7x10 ³	22.8x10 ³
Chromium and compounds	260	491	127	-	-	-	-
Copper and compounds	466	318	-	-	-	-	-
Fluorine (as HF)	246x10 ³	-	116x10 ³	55.9x10 ³	79.1x10 ³	30.8x10 ³	58.7x10 ³
Lead and compounds	344	418	-	-	-	-	-
Mercury and compounds	120	12.0	107x10 ³	209	95.0	54.7	76.6
Nickel and compounds	255	106	126	271	341	254	305
Zinc and compounds	-	-	-	265	349	220	290
PM₁₀	1.740x10⁶	0.812x10⁶	0.587x10⁶	0.394x10⁶	0.0997x10⁶	0.100x10⁶	0.286x10⁶

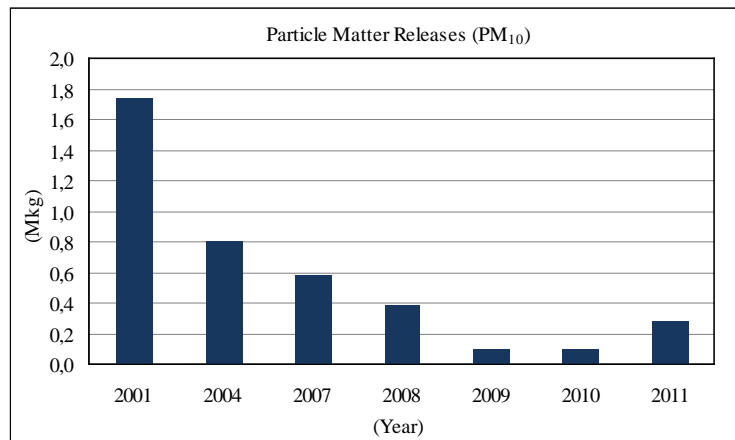


Fig. 2: Particle Matter Releases (PM₁₀) in Mkg [16].

Measurement locations

For the present study were defined two circular areas centred in stacks: one with approximately 113 km² and the other with 1 257 km². Forty-one relevant measurement locations were established within both circular areas. About sixteen urban stations were established inside a 6-km circle (A₁) around the stacks and other twenty five were established at distances from 6 to 20

km maximum from the coal plant stacks (A_2). The study area was defined considering the height of the stacks, local and detailed meteorological patterns such as wind direction, velocity and frequency; human settlements distance and the accessibility of the locations (Fig. 3) [1].

The stack's emissions influence was defined considering the possible effect on the people living in the neighborhood of the coal-fired power plant, up to 20 km, resulting from particulate matter releases, mainly from past situations. The distance of the defined measurement locations with respect to the coal plant ranged from 903.20 to 22 327.78 m.

Several measurement locations were also established in the boundary of the 20 km circle (location N° 21, 22, 25, 26, 27, 28, 30, 32, 39 and 45) primarily defined as control as theoretically they are out of influence of fly ash dispersion and the deposition is expected to be the lowest (Fig. 3).

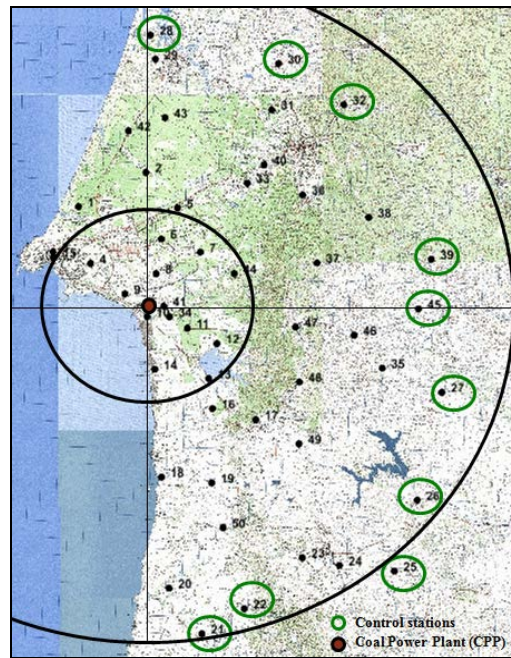


Fig. 3: Study Area and Measurement Locations (Areas: A_1 and A_2).

Field site measurements

The radiation monitoring was achieved both on and off site, being the boundary delimited by the 20 km circle centered in the stacks of the coal plant. The monitoring consisted on measuring the radionuclides concentration by gamma spectrometry at 41 measuring locations twice a year during three years. The measurements taken at off-site locations were carried out in the dominant wind direction as well as in the opposite direction and were used as control. Once the radionuclides concentrations were established in the control locations, they were compared to radionuclides concentrations at sites of interest [1].

The gamma radiation measurements were performed using a Canberra high purity coaxial detector (HPGe) based gamma spectrometer (Falcon 5000, Canberra). Energy and efficiency calibrations were done using a Eu-152 source. Genie 2000 and ISOCS (In Situ Object Counting Systems) Canberra software were used to identify and quantify the radionuclides detected in each measurement location. Counting was done for 5 000 seconds at in situ measurements as well as for the calibration. The gamma energy lines were matched at various levels to the database of possible isotopes resulting from naturally occurring radioactive materials (NORM) [1].

Doses rates determination

The absorbed dose rate in air (D) at the height of approximately 1 m above the ground was calculated assuming that the radionuclides are uniformly distributed in the ground (Eq.1) [3]:

$$D(\eta\text{Gy/h}) = 0.0417C_K + 0.462C_{Ra} + 0.604C_{Th} \quad (\text{Eq. 1})$$

where D is the absorbed dose rate (in nGy/h), C_K , C_{Ra} , and C_{Th} are the activity concentrations of K^{40} , Ra^{226} and Th^{232} , respectively (in Bq/kg).

To estimate the annual effective dose rate (E), it was used the conversion coefficient from the absorbed dose in air to the effective dose (0.7 Sv/Gy) and the outdoor occupancy factor (0.2) proposed by UNSCEAR [3] (Eq. 2):

$$E(\text{mSy/h}) = D(\eta\text{Gy/h}) \times 8760(\text{h/y}) \times 0.2 \times 0.7(\text{Sv/Gy}) \times 10^{-6} \quad (\text{Eq. 2})$$

RESULTS AND DISCUSSION

Radionuclides specific activity

The identified gamma-ray emitting radionuclides are those associated with natural background, i.e., U-238 series, Th-232 series as well as their decay daughters and K-40, and nuclear weapon tests fallout product Cs-137. Only natural radionuclides series will be of interest in the present study.

Specific software was used to identify and quantify the radionuclides detected in each measurement location (Genie 2000 and ISOCS, Canberra). Range values (Min-Max), arithmetic mean (AM), standard deviation (SD), median (MD) and geometric mean (GM) measured in the vicinity of the coal plant (0-20 km) are presented in TABLE II for the selected radionuclides: ^{40}K , ^{226}Ra and ^{232}Th . The specific activities measured in the prevailing wind direction (NW-SE) are also presented (TABLE III).

TABLE II: Activity Concentration in the Vicinity of the Coal-Fired Power Plant (in Bq/kg).

Radionuclide (Bq/kg)	Min.	Max.	AM	SD	MD	GM
K-40	62.3	795.1	361.8	177.8	353.1	314.5
Ra-226	7.7	41.3	20.4	7.9	18.3	19.0
Th-232	4.7	71.6	23.6	15.2	19.4	19.3

TABLE III: Activity Concentration in the Prevailing Wind Direction (in Bq/kg).

Radionuclide (Bq/kg)	Min.	Max.	AM	SD	MD	GM
K-40	362.8	795.1	494.7	150.3	447.7	477.4
Ra-226	22.7	41.3	29.3	6.3	28.8	28.7
Th-232	25.0	71.6	41.3	14.1	40.5	39.4

The measured radionuclides concentration in the vicinity of the coal plant ranged from 7.7 to 41.3 Bq/kg for Ra-226 and from 4.7 to 71.6 Bq/kg for Th-232, while K-40 concentrations ranged from 62.3 to 795.1 Bq/kg; the average values were 20.4, 23.6 and 361.8 Bq/kg, respectively. Higher values were registered in the prevailing wind direction as expected, due to the atmospheric dispersion with wind patterns: 362.8-795.1 Bq/kg for K-40, 22.7-41.3 Bq/kg for Ra-226 and 25-71.6 Bq/kg for Th-232. The histograms of the generic results are presented in Fig. 4 and Fig. 5 for both defined areas, A₁ and A₂, respectively.

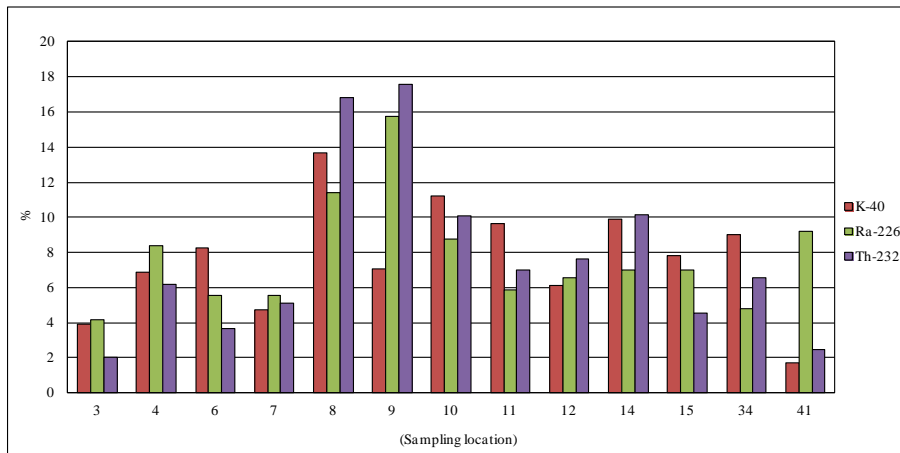


Fig. 4: Histogram for Measured Radionuclides: K-40, Ra-226, and Th-232 (A₁).

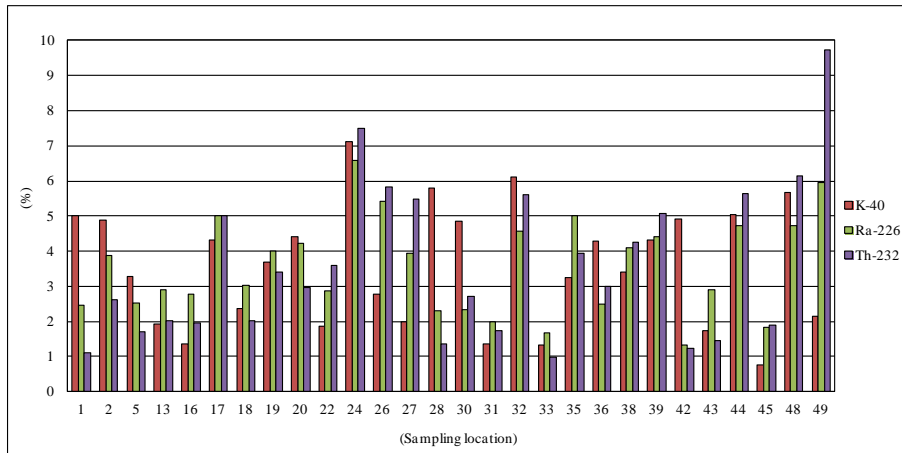


Fig. 5: Histogram for Measured Radionuclides: K-40, Ra-226, and Th-232 (A_2).

While lower radionuclides concentration values were measured at N and S from the stacks (locations N° 43 and 18, respectively) at approximately 11 km from the stacks, the highest radionuclides concentration were measured near the power plant (locations N° 8 and 9) and at distances between 6 and 20 km from the stacks, mainly in the prevailing wind direction (locations N° 17, 24, 26, 27 and 49).

Only for a few control measuring locations (N° 22, 26, 27, 45 and 49 for K-40; N° 22, 28, 30 and 45 for both Ra-226 and Th-232) the detected values were lower, as expected, for locations at 20-km boundary and theoretically away from the possible impact of the coal plant emissions.

Considering the inner area (A_1) the maximum values were measured in a few locations very close to the stacks: locations N° 8 (NE) and N° 10 (SE) for K-40; locations N° 8 (NE), N° 9 (N) and N° 10 (SE) for both Ra-226 and Th-232. On the other hand, the maximum values within the outer area (A_2) were registered at about 12.5 km from the stacks and near the boundary of 20 km: locations N° 17 (SE), 20 (SE), 24 (SE), 48 (SE) and 32 (NE) for K-40, Ra-226 and Th-232, location N° 49 (SE) for both Ra-226 and Th-232.

The measured values were interpolated by kriging using Golden Software Surfer 8 and mapped as iso-concentration contour values and are represented in Fig. 6, Fig. 7 and Fig. 8; a three dimensional distribution is also represented in these figures.

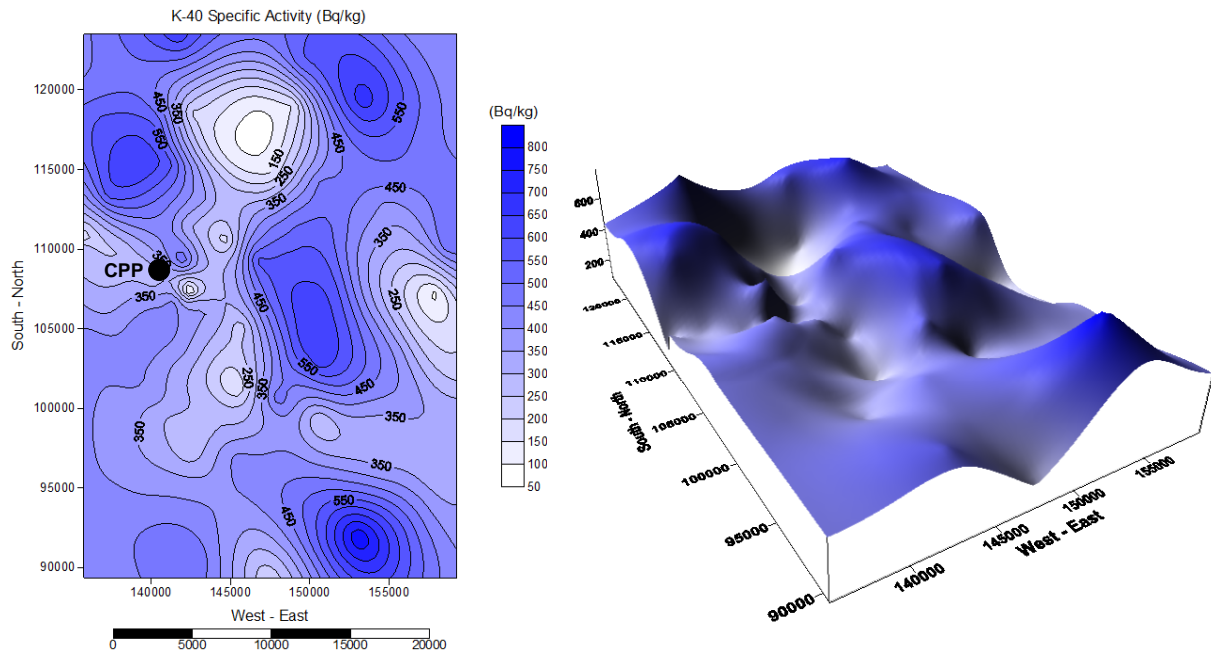


Fig. 6: Maps of Iso-Concentration Measured for K-40 (Bq/kg) (CPP: Coal-Fired Power Plant).

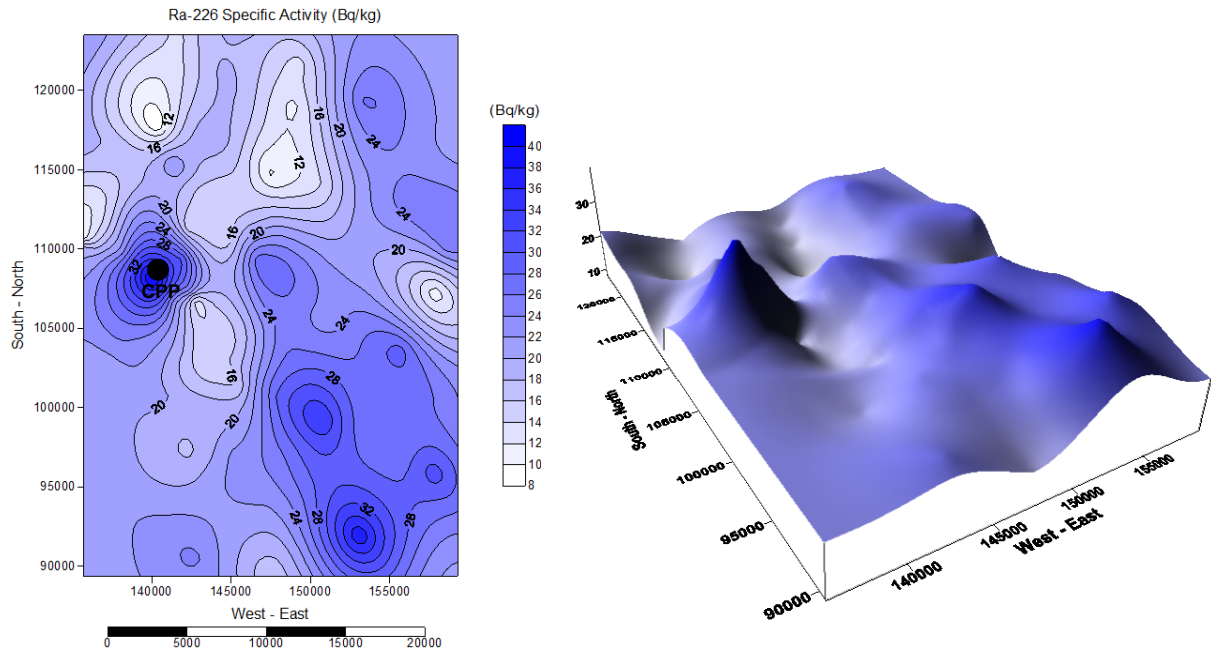


Fig. 7: Maps of Iso-Concentration Measured for Ra-226 (Bq/kg) (CPP: Coal-Fired Power Plant).

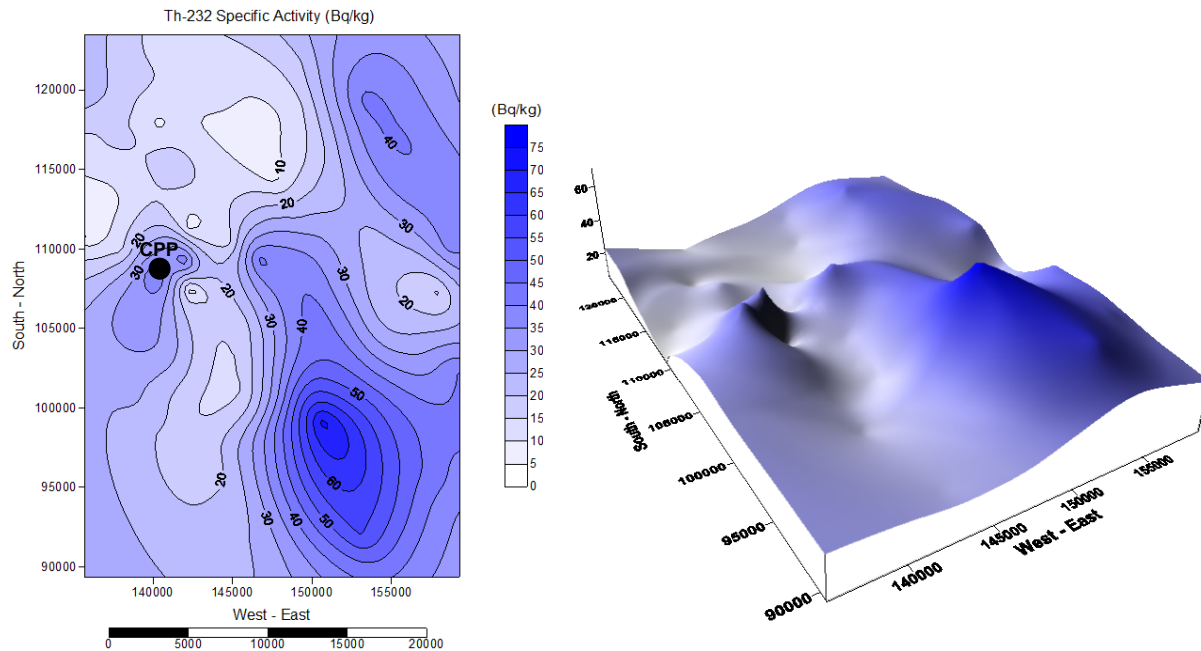


Fig. 8: Maps of Iso-Concentration Measured for Th-232 (Bq/kg) (CPP: Coal-Fired Power Plant).

The contour maps show a general trend where higher values were detected at distances between 7.5 and 20 km away from the stacks (locations N° 24, 25, 32, 46, 49 and 48) [1]. In general, radionuclides concentrations are higher at NE and SE from the stacks well beyond the distance of 6 km from the stacks.

Two hot-spots were registered within the 6 km area at locations N° 8 (3 km NW) and N° 9 (1 km NW) due to Ra-226 and Th-232 activity concentrations which may be attributed to the influence of the coal stockpiled near to the stacks (1.3 million tonnes) [1]. A similar increase in K-40 concentration activity was not observed at location N° 9.

The radionuclides concentration as a function of the distance to the power plant, mainly in the prevailing wind direction (SE), is represented in Fig. 9.

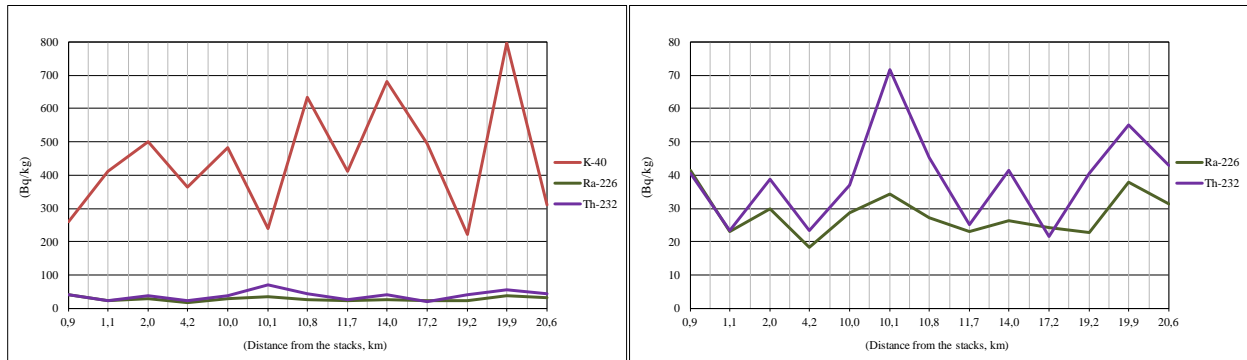


Fig. 9: Measured Concentrations of K-40, Ra-226 and Th-232 with the Downwind Distance from the Stacks (SE).

In general, higher concentrations were found nearest the power plant (0.9, 2.0 km) as well as for longer distances from 10 to 20 km from the coal plant. The concentration’s variation with the distance is very similar for Ra-226 and Th-232 with the exception for the sampling point N° 49 (10.1 km) where the Th-232 concentration is almost twice the Ra-226 concentration, in addition to the fact that K-40 presents the lowest concentration in this location. Moreover, K-40 presents a high variability along the considered direction.

Gamma dose rate

The statistical data for the absorbed gamma dose rates in air at 1 m above the ground due to the activity concentrations of Th-232 and U-238 series and K-40 are presented in TABLE IV.

TABLE IV: Absorbed Gamma Dose Rates in Air at 1 m Above the Ground (η Gy/h).

	K-40 (η Gy/h)	Ra-226 series (η Gy/h)	Th-232 series (η Gy/h)	Total dose rate (η Gy/h)
Min.	2.60	3.55	2.85	13.97
Max.	33.16	19.08	43.22	84.00
AM	15.09	9.44	14.25	38.78
SD	7.41	3.65	9.15	16.64
MD	14.73	8.47	11.69	37.82
GM	13.11	8.78	11.66	35.33

The total absorbed gamma dose rates in air at 1 m above the ground, due to the activity concentrations of K-40, Ra-226 and Th-232 series, ranged from 2.60-33.16 η Gy/h, 3.55-19.08 and 2.85-43.22 η Gy/h, respectively. The total absorbed dose rates ranged from 13.97-84.00 η Gy/h with an average value of 38.78 η Gy/h.

The worldwide average dose rates values ranges from 18 to 93 η Gy/h and the population-weighted average absorbed dose rate is approximately 60 η Gy/h [3]. The average value obtained in this study is lower than the population-weighted worldwide average. However, this value was highly exceeded in eight measuring locations: N° 8, 9, 17, 24, 32, 44, 48 and 49. Moreover,

measurements from previous studies carried out in the same region in 1993, registered values in the range of 17-78 $\eta\text{Gy/h}$ with an average value of 41 $\eta\text{Gy/h}$ [17]. This average value was also exceeded in 16 locations: N° 2, 8, 9, 10, 17, 19, 20, 24, 26, 32, 35, 38, 39, 44, 48, 49 (TABLE V).

TABLE V: Total Absorbed Gamma Dose Rates in Some Measurements Locations.

N°	Total dose ($\eta\text{Gy/h}$)	Direction	N°	Total dose ($\eta\text{Gy/h}$)	Direction
2	44.76	N	10	41.90	SE
8	58.19	N	17	55.74	SE
9	54.40	N	19	42.98	SE
20	45.01	S	24	84.00	SE
32	65.54	NE	26	53.33	SE
38	45.60	NE	35	46.01	SE
39	54.34	NE	48	66.38	SE
44	61.18	NE	49	69.10	SE

All data set of the total absorbed dose rates due to the activity concentrations of Th-232, Ra-226 and K-40 were plotted as an isodose contour map as presented in Fig. 10. The three-dimensional dose rate distribution is also presented in the same figure.

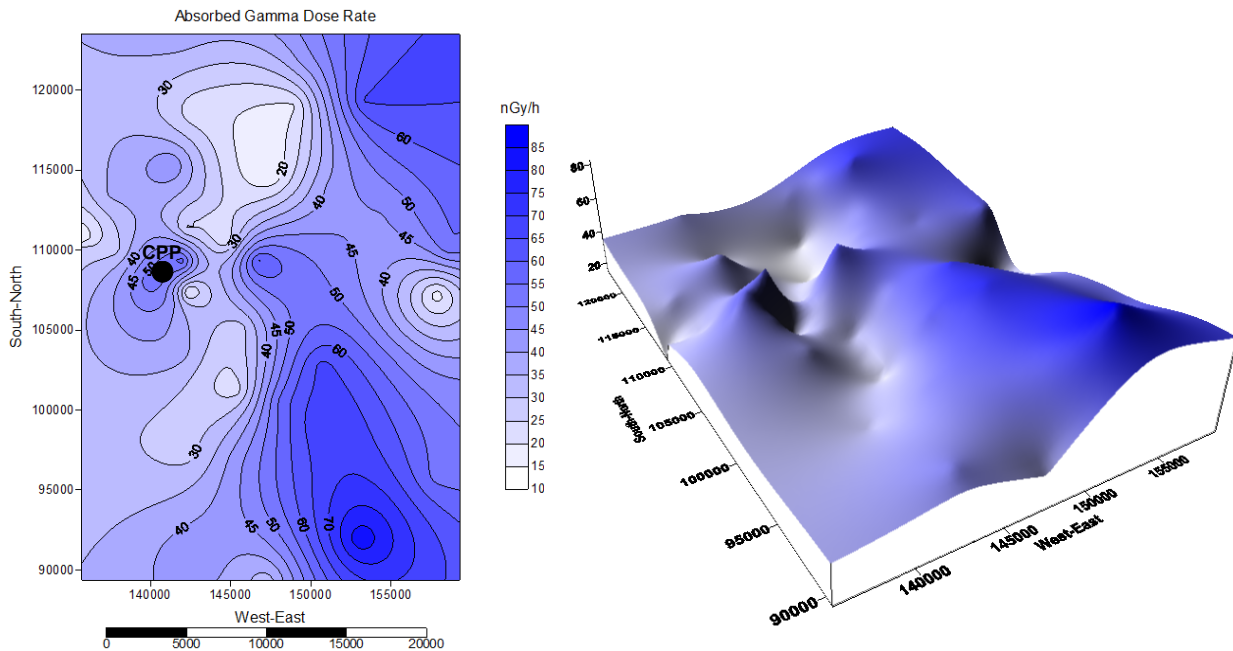


Fig. 10: Isodose Map of the Total Absorbed Gamma Dose Rate (CPP: Coal-Fired Power Plant).

The highest values occur near the stacks and at distances between 10 and 20 km to NE and SE from the stacks. The lowest doses are found between these two regions.

The percentage contribution to the total absorbed gamma dose rate due to K-40, Ra-226 and Th-232 concentration at each measurement location is presented in Fig. 11.

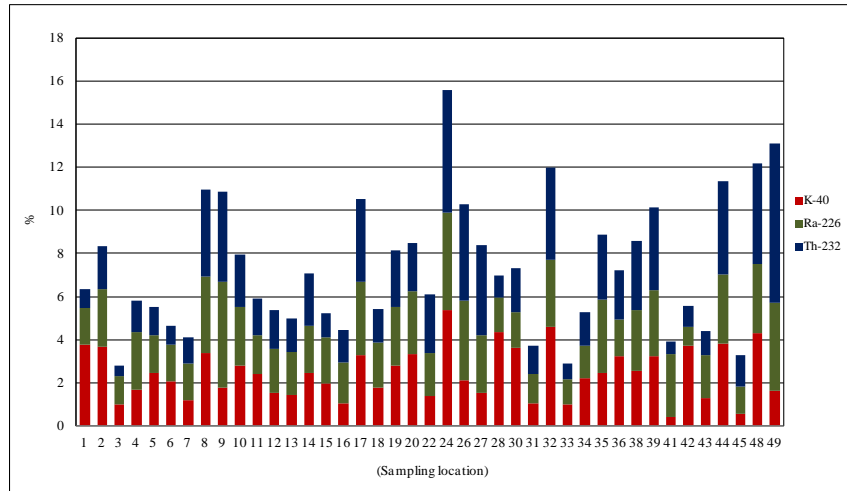


Fig. 11: Percentage Contribution to the Total Dose Rate due to K-40, Ra-226 and Th-232 Concentration.

The major contribution to the total absorbed dose comes from joint K-40 and Th-232 activity concentrations. Only at location N° 43 the highest contribution to total absorbed dose comes from Ra-226 activity concentration. Total absorbed dose rates observed at locations N.º 8, 9, 17, 24, 26, 44, 48 and 49 (all at SE from the stacks) show the highest values due to Th-232 contribution while other locations: N° 3, 4, 15 (W-SE); 1 (SW-N); 2, 5, 6, 28, 33, 30 (N-NE); 7, 32, 36 (NE-E) and N° 10, 11, 14, 18, 19, 20 and 34 (S-SE) show higher dose rate values due to the K-40 contribution.

The gamma radiation dose rates are higher than the average reported by [17] for this region: 37.6 to 41.5 nGy/h or the Portuguese average, as reported by UNSCEAR 2000 [3]: 84 nGy/h ranging from 4 to 230 nGy/h.

The relative contribution to the total absorbed dose rate due to K-40, Ra-226 and Th-232 activity concentration is presented in Fig. 12.

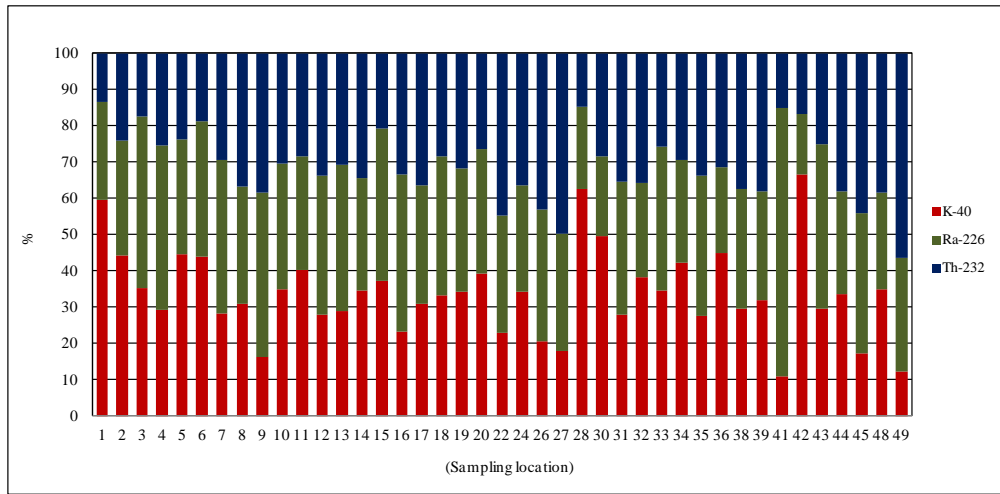


Fig. 12: Relative Contribution to the Total Dose Rate due to K-40, Ra-226 and Th-232 Concentration.

The estimated relative contribution to the total dose rate by the individual components of natural radioactivity is in average 34% from K-40, 35% from Ra-226 and 31% from Th-232.

Annual dose rate

The outdoor effective dose rate was calculated according to Eq. 2. The values ranged from 0.017 to 0.103 mSv/y with an average value of 0.048 mSv/y (Fig. 13) which is significantly lower than the worldwide average exposure of 0.07 mSv/y and within the exposure level recommended for the public of 1 mSv/y [18]. This value was exceeded at 6 locations at SE from the stacks (N° 8, 24, 32, 44, 48, 49). The highest and lowest effective doses were registered at measurement locations N° 24 (SE) and N° 3 (NW), respectively.

Studies conducted in the vicinity regions of the considered site have shown that the annual effective outdoor dose is, in average, 0.05 and 0.06 mS/y [17].

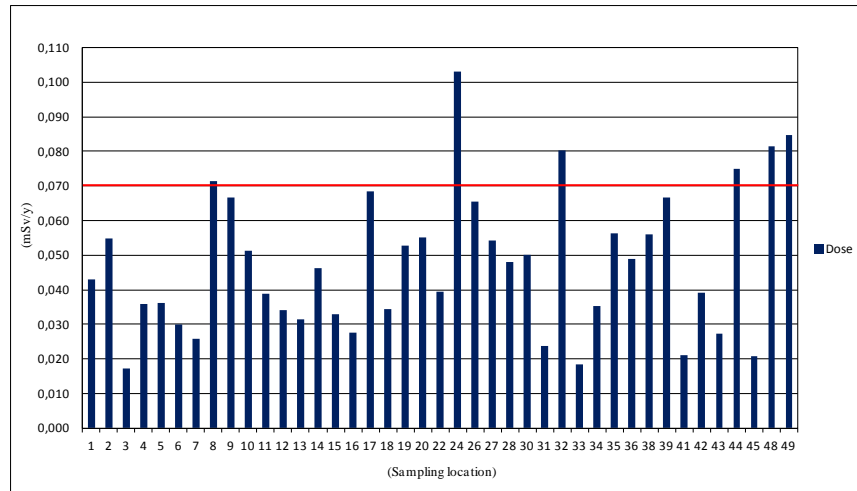


Fig. 13: Outdoor Effective Dose Rate (mSv/y).

CONCLUSIONS

The environmental monitoring of the natural background radiation was carried out in the surroundings of a Portuguese coal-fired power plant.

The average radionuclides concentration of K-40, Ra-226 and Th-232 is 361.8 ± 177.8 Bq/kg, 20.4 ± 7.9 Bq/kg and 23.6 ± 15.2 Bq/kg, respectively. In general, the measured values are similar to those presented in the literature, as background levels. However, these average values are increased in 37% for K-40, 44% for Ra-226 and 75% for Th-232 in measurements carried out in the prevailing wind direction (SE). Radionuclides concentrations with the distance to the coal plant in the prevailing wind direction are not uniform but the generic trend reflects an increasing with the distance; an enrichment of the radionuclides activity concentrations could be observed with the distance to the coal plant up to a distance of 20 km. The activity concentrations of Ra-226 and Th-232 showed a good correlation (0.83) for each measurement location while a weak correlation was observed between each one of the previous radionuclides and K-40 (0.31 and 0.39, respectively).

The total absorbed gamma dose rate is, in average, 38.78 ± 16.64 η Gy/h and in the prevailing wind direction is 61.78 η Gy/h where the highest value was found to be 84.00 η Gy/h. The obtained average dose rate is lower than the world average value of 60 η Gy/h, suggested by UNSCEAR [3], and the average value from previous studies developed in the region, 41 η Gy/h.

The percentage contribution to the absorbed gamma dose rate is, in average 34% from K-40, 35% from Ra-226 and 31% from Th-232. However, for those measurements locations where the highest dose rates were registered, the major contribution comes from Th-232 (contribution between 36% and 56%). This indicates that the major contributor to gamma dose rate in the

prevailing wind direction is Th-232.

In general, the highest values were obtained in two locations near the stack considered to be two hot spots, and in several locations in the dominant wind direction at distances between 6 and 20 km from the stacks. The hot spots present different nuclide contributions to the absorbed gamma dose: in location N° 8: 30.79% K-40, 32.57% Ra-226, 36.63% Th-232 and in location N° 9: 16% K-40, 45.32% Ra-226, 38.57 Th-232. These locations are not in the line with the prevailing wind direction which suggests that the coal piles near the stacks without any protection against climate agents may be the origin of unburned coal particles dispersed and accumulated in these locations. The ashes disposal may also be a contributing source.

The outdoor effective dose rate is, in average, 0.048 ± 0.0204 mSv/y, and this value is lower than the world average outdoor value of 0.07 mSv/y. However, the obtained values were higher than this limit for some locations at SE from the coal plant. It was possible to observe an increase in environmental radiation of natural radionuclides and corresponding dose rates which can be attributed to the past emissions in the previous decades. Nevertheless, due to the relatively low natural levels of these radionuclides, the annual average effective dose is lower than 0.07 mSv/y.

In general, the environmental radiation monitoring carried out in the vicinity of the studied coal plant did not show outstandingly high values. However, significantly high differences were measured near the stacks and mostly in the prevailing wind direction locations at distances up to 20 km from the stacks. In these cases absorbed gamma dose rate and effective outdoor dose rate limits were exceeded.

Radioactive nuclides as K-40, Ra-226 and Th-232 escapes from the stacks of coal-fired power plant in gaseous or particulate form, disperse into the atmosphere and deposit on the ground but only 6% of the emitted particles from a coal power plant with a 300 m stack fell within the distance of 5 km [19]. For power plants equipped with electrostatic precipitators most particles emitted have less than 2 μm in diameter which can agglomerate accelerating the deposition near the source but even though, smaller particles can travel between 100 and 1700 km [19]. The height of the stacks (225 m) combined with wind patterns (predominantly from NW to SE) are expected to have transported the emissions up to long distances away from the stacks explaining the higher values up to 20 km from the stacks. In this case, the peak values detected near the stacks could not have fallen from the plume vertically.

REFERENCES

1. M. L. DINIS, A. FIÚZA, J. S. DE CARVALHO, J. GÓIS and A. C. M. CASTRO, “Radiological impact associated to technologically enhanced naturally occurring radioactive materials (TENORM) from coal-fired power plants emissions”. Proceedings of the

- “International Conference Waste Management Symposia (WMS), Phoenix, USA, February 24-28 (2013).
2. UNSCEAR 1982, “*Sources and biological effects*”. United Nation Scientific Committee of the Effect Atomic, Radiation Report to The General Assembly, with annexes. United Nation sales, publication E.82.IX.8., United Nations, New York (1982).
 3. UNSCEAR 2000, “*Sources and effect of ionizing radiation*”. United Nation Scientific Committee of the Effect Atomic, Radiation Report on The General Assembly, United Nation, New York (2000).
 4. H. L. BECK, “*Radiation exposures due to fossil fuel combustion*”. *Radiat. Phys. Chem.* 34 (2), 285–293 (1989).
 5. C. PAPASTEFANO, “*Radiation impact from lignite burning due to 226Ra in Greek coal-fired power plants*”. *Health Phys.* 70 (2), 187–191 (1996).
 6. R. DELFANTI, C. PAPUCCI, C. BENCOB, “*Mosses as indicators of radioactivity deposition around a coal-fired power station*”. *Sci. Total Environ.* 227, 49–56 (1999).
 7. A. BABA, “*Assessment of radioactive contaminants in by-products from Yatagan (Mugla, Turkey) coal-fired power plant*”. *Environ. Geol.* 41, 916–921 (2002).
 8. H. BEM, P. WIECZORKOWSKIA, M. BUDZANOWSKI, “*Evaluation of technologically enhanced natural radiation near the coal-fired power plants in the Lodz region of Poland*”. *J. Environ. Radioactiv.* 61, 191–201 (2002).
 9. M. FLUES, V. MORAES, B.P. MAZZILLI, “*The influence of a coal-fired power plant operation on radionuclide concentrations in soil*”. *J. Environ. Radioactiv.* 63, 285–294 (2002).
 10. F. ADROVIC, M. PROKIC, M.M. NINCOVIC and N. GLISSIC, “*Measurements of environmental background radiation at location of coal-fired power plants*”. *Radiation Protection Dosimetry*, Vol. 112, No. 3, pp. 439-442 (2004).
 11. A. BAEZA, J. A. CORBACHO, J. GUILLÉN, A. SALAS, J. C. MORA, B. ROBLES, D. CANCIO, “*Enhancement of natural radionuclides in the surroundings of the four largest coal-fired power plants in Spain*”. *Journal of Environmental Monitoring*, 14, pp. 1064-1072, <http://dx.doi.org/10.1039/C2EM10991C> (2011).
 12. E. CHARRO, V. PENA, “*Environmental impact of natural radionuclides from a coal-fired power plant in Spain*”. *Radiation Protection Dosimetry*, <http://dx.doi.org/10.1093/rpd/ncs126> (2012).
 13. E. CHARRO, R. PARDO, V. PENA, “*Statistical analysis of the spatial distribution of radionuclides in soils around a coal-fired plant in Spain*,” *J. Environ. Radioactiv.*, 124, pp. 84-92 (2013).
 14. M. L. DINIS, A. FIÚZA, J. GÓIS, J. S. DE CARVALHO and A. C. M. CASTRO, “*Assessment of direct radiological risk and indirect associated toxic risks originated by Coal-Fired Power Plants*”. *Radioprotection Journal*, 46, n° 6: S137-S143 (2011).

15. EDP, DECLARAÇÃO AMBIENTAL 2011, “*Central Termoelétrica de Sines*”.
<http://www.apambiente.pt/zdata/Instrumentos/GestaoAmbiental/EMAS/DA/99/11.pd>
(2011).
16. E-PRTR, “*European Pollutant Release and Transfer Register Database*”,
<http://prtr.ec.europa.eu/Home.aspx> (2010).
17. E. M. AMARAL, J. G. ALVES and V. CARNEIRO, “*Doses to the Portuguese Population due to Natural Gamma Radiation*”. *Radiation Protection Dosimetry*, Vol. 45 N°1/4 pp.541-543 (1992).
18. IAEA, 1196, “*International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of the Radiation Sources*”, IAEA, Vienna.
19. T. J. KEEGAN, M. E. FARAGO, I. THORNTON, B. HONG, R. N. COLVILE, B. PESCH, P. JAKUBIS and M. J. NIEUWENHUIJSEN, “*Dispersion of As and selected heavy metals around a coal-burning power station in central Slovakia*”. *Sci Total Environ.* 358(1-3):61-71 (2006).

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