

Radiological Impact Associated to Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) from Coal-Fired Power Plants Emissions – 13436

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ABSTRACT

Certain materials used and produced in a wide range of non-nuclear industries contain enhanced activity concentrations of natural radionuclides. In particular, electricity production from coal is one of the major sources of increased human exposure to naturally occurring radioactive materials. A methodology was developed to assess the radiological impact due to natural radiation background. The developed research was applied to a specific case study, the Sines coal-fired power plant, located in the southwest coastline of Portugal.

Gamma radiation measurements were carried out with two different instruments: a sodium iodide scintillation detector counter (SPP2 NF, Saphymo) and a gamma ray spectrometer with energy discrimination (Falcon 5000, Canberra). Two circular survey areas were defined within 20 km of the power plant. Forty relevant measurements points were established within the sampling area: 15 urban and 25 suburban locations. Additionally, ten more measurements points were defined, mostly at the 20-km area. The registered gamma radiation varies from 20 to 98.33 counts per seconds (c.p.s.) corresponding to an external gamma exposure rate variable between 87.70 and 431.19 nGy/h. The highest values were measured at locations near the power plant and those located in an area within the 6 and 20 km from the stacks. In situ gamma radiation measurements with energy discrimination identified natural emitting nuclides as well as their decay products (Pb-212, Pb-214, Ra-226, Th-232, Ac-228, Th-234, Pa-234, U-235, etc.).

According to the results, an influence from the stacks emissions has been identified both qualitatively and quantitatively. The developed methodology accomplished the lack of data in what concerns to radiation rate in the vicinity of Sines coal-fired power plant and consequently the resulting exposure to the nearby population.

INTRODUCTION

Radioactivity dispersed in the environment results not only from nuclear fuel cycle activities but also from other several technological activities. The most relevant non-nuclear industries, concerning radioactive elements emissions, are phosphates industry, ceramic industry and energy production activities. In particular, energy production from coal-fired power plants is one of the major sources of

increased exposure to man from enhanced naturally occurring materials.

Coal contains trace quantities of the naturally occurring radionuclides like uranium (1 to 10 mg/kg) and thorium (3 to 25 mg/kg), as well as their radioactive decay products and K-40.

Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) are generated when burning removes organic constituents, leaving minerals and concentrating trace quantities of naturally occurring radionuclides (uranium, thorium, potassium) and their radioactive decay products, including radium, in ash. The amount of Ra-226 in coal can vary by more than two orders of magnitude depending on the type of coal (composition in organic matter) and where it was mined.

In addition to TENORM, coal ash contains silicon, aluminium, iron, and calcium; these elements make up about 80 to 90% of all of the constituents of coal ash.

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%). Usually, part of the fly ash generated is sold to other industrial consumers when their characteristics are in agreement with legal requirements.

Fly ash is the finest portion of the coal ash particles and it is transported from the combustion chamber by exhaust gases. Stack filtration systems, such as electrostatic precipitators, baghouses, and scrubbers are routinely used to reduce the emission of fly ash to the atmosphere by at least 95 percent. A small fraction of the fly ash produced, typically 2-5 percent, is released into the air. Modern coal-fired power plants equipped with sophisticated retention devices release less than 1% of fly ash. Nevertheless, when burning huge amounts of coal per year (3 million t), 1% can be a significant amount of fly ash released with enhances radionuclide concentration.

The activities of natural radionuclide concentration released into the atmosphere from a coal power plant will depend on many factors such as the activity concentration 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 [1].

People living near coal power plants may be exposed to increasing quantities of radioactive isotopes through inhalation, external irradiation and ingestion following deposition of activity on the ground. Because 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.

This work describes the methodology developed to assess the radiological impact due to natural radiation background increasing levels, potentially resulting from the dispersion of radioactive elements present in the coal ash atmospheric emissions. The current investigation is part of a research project that is undergoing until 2013 and it was applied to a specific case study, the Sines coal-fired power plant located in the southwest coastline of Portugal. Environmental radiation monitoring in the vicinity of this coal-fired power plant was used primarily to define the variability in measured background exposures to determine the plant's contribution, both qualitatively and quantitatively, on the background radiation level.

METHODOLOGY

Study area

The present study was conducted in the vicinity of Sines coal-fired power plant located in the southwest coastline of Portugal, at 6 km south-east from the city of Sines and about 150 km south of the country's capital, Lisbon (Fig. 1). It is located in an industrial area with more than 14 other industrial plants, including a petrochemical complex, a refinery and a resins plant.



Fig. 1: Sines coal-fired power plant location (national, regional and local).

The coal-fired power plant is a conventional one, working with 4 coal fired units, each one with an electrical energy output of 314 MW [2]. The first unit started working in 1985 and the last one in 1989. The power plant operates with efficiency in the range of 32 to 35%. The plant has two operational stacks with 225 m height.

Sines coal-fired power plant consumes as primary fuel bituminous coal, imported from many different countries [3]. Coal is stored at open air in 4 active piles with 150 000 t each and in one passive pile with 700 000 t. In September of 2011, the coal power plant was burning coal with a caloric value between 5.6 and 7.2 kJ and ash content between 8.3 and 9.2%.

In 2011 the energy produced in this coal power plant was approximately 7 432 GWh with a coal consumption of 2 636 539 t (355 t/GWh); approximately 286 t of particular matter were released into the atmosphere, 39 768 t of non-conformity fly ash was conducted to disposal into the coal plant landfill as well as 26 378 t of bottom slag. At the end of 2011, the amount of accumulated material (fly ash and bottom slag) in the coal plant disposal was 1 089 771 t, covering an area of about 11 ha [4].

Data on this particular source emission is reported in the European database “European Pollutant Release and Transfer Register” [5] for 2007, 2008, 2009 and 2010 (Table I). The data refers to PM₁₀ and heavy metals releases into air. There is no information about PM_{2.5} releases in this database.

Fifteen urban stations were established inside a 6-km circle around the stacks and other twenty five were established at distances from 6 to 20 km maximum from the coal plant stacks. Later, ten more measurement stations were established, mostly in the outer area (6-20 km), in order to achieve a complete spatial distribution of the results (Fig. 2).

The measurement locations were defined taking into consideration the height of the stacks; local meteorological conditions such as wind direction, velocity and frequency; human settlements distance and the accessibility of the locations. The dominant wind direction in this region is from northwest to southwest.

A few measurement stations were selected as control: 21, 22, 25, 26, 27, 28, 30, 32, 39 and 45, located at a distance of about 20 km from the stacks plant where the influence of fly ash dispersion and deposition is expected to be the lowest.

Gamma radiation survey and radionuclides concentration

The in situ gamma radiation measurements were carried out in the surroundings of the coal plant considering the possible enhance in the background radiation up to distances of 20 km from fly ash stacks emissions, mainly due to past situations. The measurements were carried out in different seasons: at the end of summer (September 2011) and in the beginning of spring (March 2012) at the same measurements locations. The measurements were performed by both scintillation and gamma-ray spectrometry.

The radiation survey was conducted using a sodium iodide scintillation detector counter (SPP2 NF, Saphymo) at 1 meter above the ground. Five measurements were done at each location. The values were plotted as a contour map by using software surfer.

A few measurements were also carried out outside the 20-km circle in the dominant wind direction as well as in the opposite direction to establish the background situation. Once background radiation count rates levels were established, they were compared to count rates at sites of interest.

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 station. Counting was done for 5 000 seconds in situ measurements as well as for the calibration. The gamma energy lines were matched at various levels to a library of possible isotopes resulting from naturally occurring radioactive materials (NORM).

RESULTS AND DISCUSSION

Gamma radiation survey

The results of gamma radiation obtained with the scintillation counter SPP2-NF (c.p.s.) are presented in TABLE II for two survey campaigns (September 2011 and March 2012). Range values (Min-Max), arithmetic mean (AM), standard deviation (SD), median (MD) and geometric mean (GM) are presented

for both sampling campaign, as well as the correspondent exposure dose rate (nGy/h).

TABLE II: Gamma Radiation Measurements with the Scintillation Counter (SPP2-NF, Saphymo).

	γ - radiation (c.p.s.)		Dose rate (nGy/h)	
	September 2011	March 2012	September 2011	March 2012
Min.	20.00	20.83	87.70	91.35
Max.	96.00	98.33	420.96	431.19
AM \pm SD	52.47 \pm 22.35	52.05 \pm 21.01	230.08 \pm 98.01	228.22 \pm 82.13
MD	49.50	50.42	217.06	221.07
GM	47.74	47.78	209.33	209.53

The measured gamma radiation range from 20 to 98.33 c.p.s. and the exposure rate range from 87.70 to 431.19 nGy/h (0.01 mR/h to 0.049 mR/h)¹ for both survey campaigns.

The arithmetic mean values for first and second campaigns were (52.47 \pm 22.35) and (52.05 \pm 21.01) c.p.s., respectively. Measured values are similar for both campaigns; a seasonal variation could not be observed.

The gamma radiation dose rates are higher than the average reported by [6] for this region: 37.6 to 41.5 nGy/h or the Portuguese average as reported by UNSCEAR 2000 [7]: 84 nGy/h ranging from 4 to 230 nGy/h.

The gamma radiation rates (c.p.s.) obtained during the 1st and 2nd survey campaigns (September 2011; March 2012, respectively) were plotted as contour maps. The results are presented in Fig. 3.

¹ Considering that 1 roentgen (R) of air kerma deposits 0.00877 gray of absorbed dose in dry air [8].

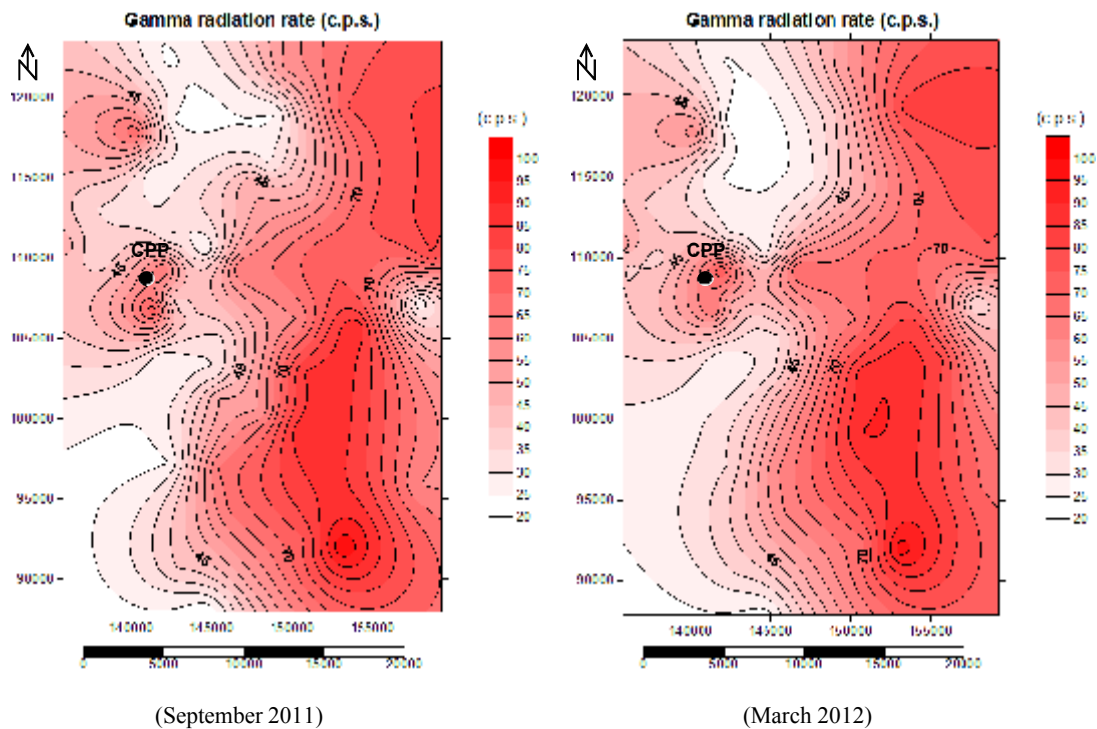


Fig. 3: Maps of iso-values of gamma radiation for both survey campaigns (CPP: Coal Power Plant).

Both contour maps are quite similar: higher values were detected at distances between 7.5 and 20 km away from the stacks (stations 24, 25, 32, 46, 49 and 48).

The lowest values were measured at stations 21 and 43 located at S and N of the stacks. Only for a few control stations (21, 27, 28 and 45) the detected values were low as expected for locations at 20-km boundary and “theoretically” away from the possible impact of the coal plant emissions.

Maximum values within the 6 km area corresponds to two stations close to the stacks (stations 8 and 10) located at NW and E from the stacks. Maximum values within the 6-20 km area were detected at stations located at about 12.5 km from the stacks and for stations located near the boundary of 20 km (25, 24, 48, 49 at SE and 32 at NE).

Two hot spots stand out at two locations very near to the stacks (8 and 10) in both survey campaigns, as shown in the following figure (Fig. 4).

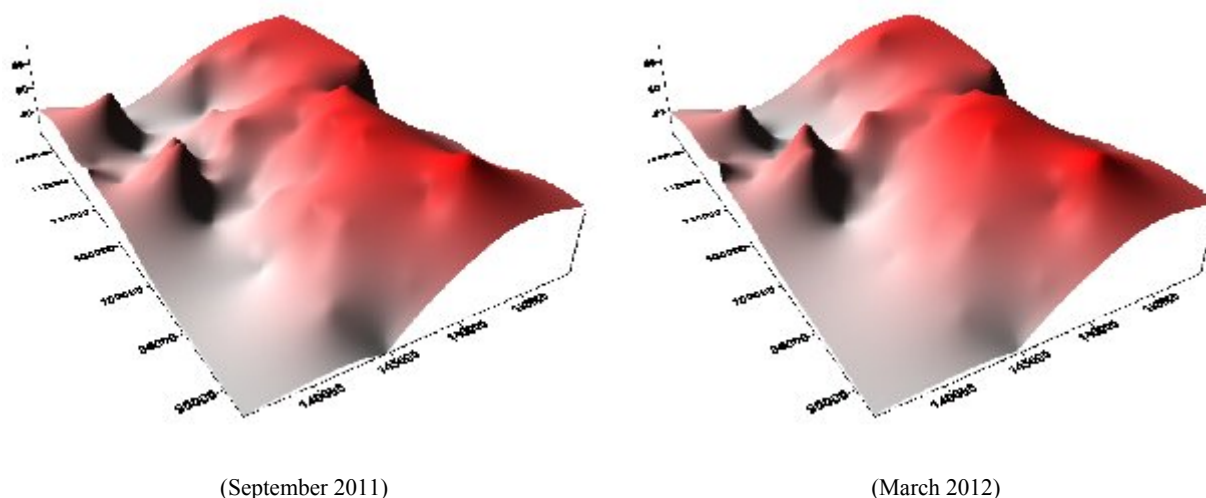


Fig. 4: Three-dimensional representation of gamma radiation rates (c.p.s.) for both survey campaigns.

One of the hotspots is located at east from the stacks while the other is located at north. The first one may be attributed to the influence of the coal stockpiled near to the stacks (1.3 million tonnes), while the other it is suspected to be related with a streamlet where the effluents from the coal stockpile, dust extractors and others cleaning activities are discharged, after previous treatment. This should be sustained by further research in the near future (water and sediments analysis).

In order to establish a relation between the obtained results and the wind patterns (direction and velocity) the survey area was divided in 45° sectors and the results were compiled according to their location in each one of the defined sectors.

Range values (Min-Max), arithmetic mean (AM), standard deviation (SD), median (MD) and geometric mean (GM) of gamma radiation rate (c.p.s.) are presented in TABLE III and TABLE IV as well as the corresponding exposure dose rate (nGy/h), for both campaigns.

TABLE III: Gamma Radiation and Exposure Dose Rates for Each Sector (1st survey campaign).

	NW-N		N-NE		NE-E		E-SE		SE-S	
	(c.p.s.)	(nGy/h)	(c.p.s.)	(nGy/h)	(c.p.s.)	(nGy/h)	(c.p.s.)	(nGy/h)	(c.p.s.)	(nGy/h)
Min.	38.00	166.63	20.83	91.35	27.00	118.40	26.00	114.01	20.00	87.70
Max.	55.00	241.18	95.00	416.58	90.00	394.65	90.00	394.65	96.00	420.96
AM	47.40	207.85	40.99	156.40	60.87	266.92	55.83	244.83	53.30	233.72
SD	6.23	27.31	19.47	88.66	21.38	93.77	23.33	102.32	28.00	122.78
MD	48.00	210.48	34.00	149.09	70.83	310.60	60.00	263.10	46.00	201.71
GM	47.06	206.34	36.93	161.92	56.94	249.67	51.09	224.02	47.67	209.04

For the first campaign the lowest values (38.00 c.p.s.) were measured at NW-N sector and the highest values (96.00 c.p.s.) were measured at the SE-S sector, although for N-NE, NE-E and E-SE sectors the values were high as well: 95.00, 90.00 and 90.00 (c.p.s.), respectively.

TABLE IV: Gamma Radiation and Exposure Dose Rates for Each Sector (2nd survey campaign).

	NW-N		N-NE		NE-E		E-SE		SE-S	
	(c.p.s.)	(mR/yr)	(c.p.s.)	(mR/yr)	(c.p.s.)	(mR/yr)	(c.p.s.)	(mR/yr)	(c.p.s.)	(mR/yr)
Min.	38.33	168.09	20.83	90.63	25.83	113.28	34.17	149.82	22.50	98.66
Max.	57.50	252.14	75.00	326.25	81.67	358.11	90.00	394.65	98.33	431.19
AM	46.00	201.71	38.22	161.01	58.98	258.63	60.37	264.73	52.80	231.54
SD	9.10	39.90	16.40	71.33	19.59	85.89	21.68	95.05	23.54	103.20
MD	40.83	179.05	30.00	130.05	54.17	310.60	65.00	285.03	48.33	211.94
GM	45.31	198.68	34.32	149.28	53.81	243.08	56.57	248.04	48.00	210.48

For the second campaign the same variation pattern was observed: the lowest values (38.33 c.p.s.) were measured at NW-N sector and the highest values (98.33 c.p.s.) at the SE-S sector, although for the E-SE sector the values were high as well (90.00 c.p.s.).

For the measurements carried out outside the 20-km circle, the highest values (85 c.p.s.) were obtained in the dominant wind direction (SE) and the lowest values (20 c.p.s.) were obtained in opposite dominant wind direction (NW-N). The average value in this direction, outside the 20-km boundary, was 25 c.p.s. (109.62 nGy/h) and this was the value established for the background.

Radionuclides activity concentration identified by γ -spectrometry

In situ gamma radiation measurements identified natural emitting nuclides, as well as their decay products, like uranium and thorium decay chain elements. Nevertheless, some unidentified peaks still need to be identified. Some radionuclides were not identified in all stations like Bi-211, Bi-214, Ra-224 and Ra-226, Tl-208 and Ac-228 (Table V); the results referring to these radionuclides should be interpreted with some reserves as data from posterior surveys carried out in September and October of 2012 will fill this gap.

TABLE V: Measured Activity Concentration by γ -Spectrometry in the Vicinity of the Coal Power Plant.

Nuclide	²³⁵ U series (Bq/kg)	²³⁸ U series (Bq/kg)			²³² Th series (Bq/kg)				⁴⁰ K (Bq/kg)	
	Bi-211	Bi-214	Pb-214	Ra-226	Tl-208	Bi-212	Pb-212	Ra-224	Ac-228	K-40
N ²	18	23	38	16	25	36	35	11	23	40
Min.	0.93	11.83	5.82	26.71	1.82	5.47	7.03	4.47	2.24	84.14
Max.	73.68	61.19	34.30	69.70	28.18	49.81	46.91	54.23	72.84	1895.07
AM	25.15	29.39	17.95	47.02	9.61	21.72	23.04	21.62	29.88	457.58
SD (\pm)	23.06	13.98	7.00	13.87	5.95	11.93	12.44	15.68	15.84	349.46
GM	12.42	26.56	16.60	44.02	8.14	18.55	19.99	16.50	16.89	366.97

For those radionuclides identified and quantified in almost every measurement stations (Pb-214, Pb-212 and Bi-212), the values vary over a relatively large range, from 5.82 to 49.81 Bq/kg. For K-40 the

² N: Number of measurements stations where the respective radionuclide was identified.

variability is also very significant: from 84.14 to 1895.07 Bq/kg. An iso-concentration map was drawn for the measurements of both sampling campaigns (Fig. 5).

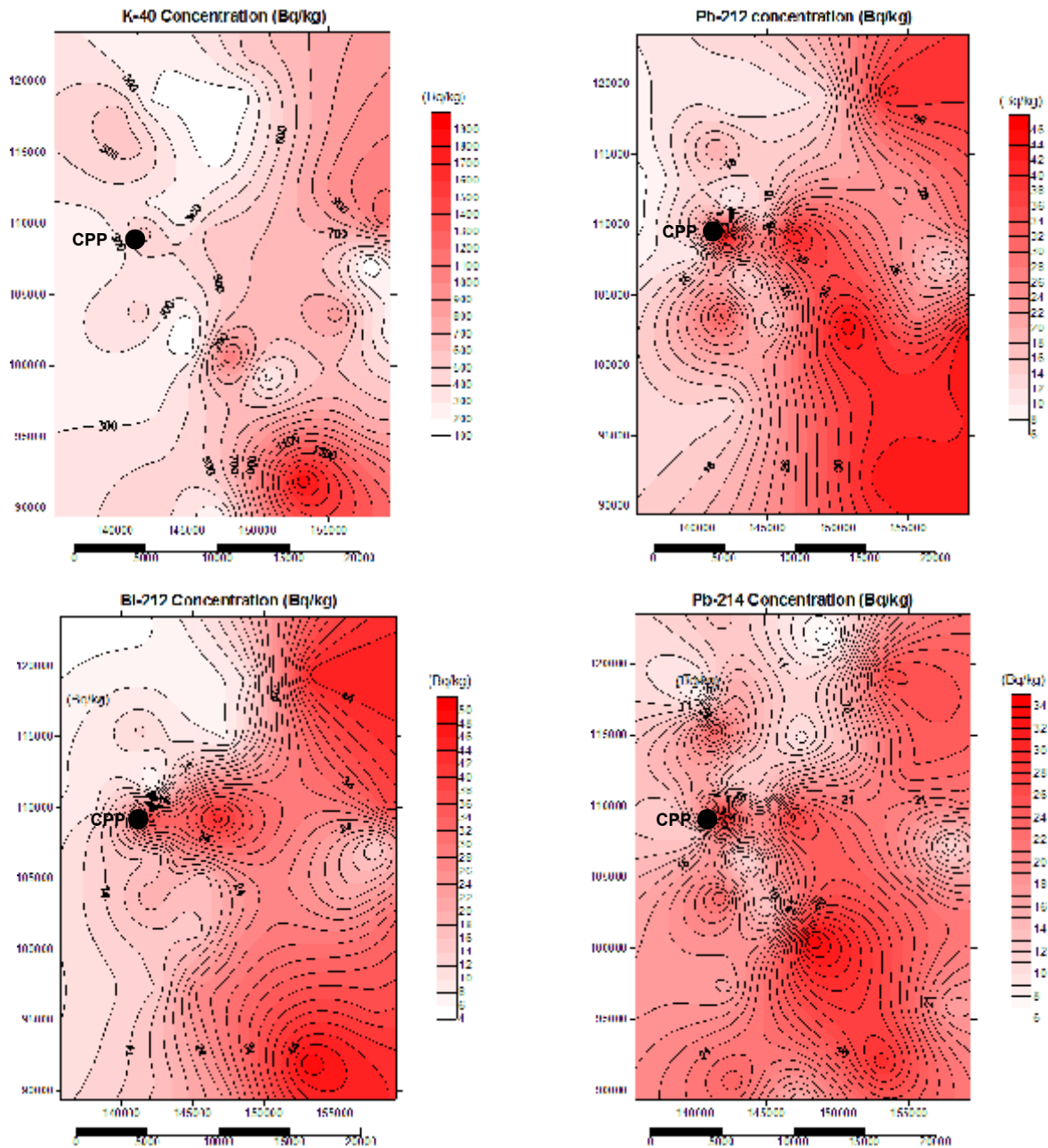


Fig. 5: Maps of iso-concentration measured for K-40, Pb-212, Bi-212 and Pb-214.

The results show that the highest values occur to distances away from the stacks, with a few exceptions (the same hotspots, stations 8 and 10), which are in a good agreement with the scintillation counter (SPP2) as it is possible to observe the same spatial pattern.

In table VI it is presented the relation between the maximum values obtained for each one of the identified radionuclides and the distance to the stacks as well as the direction.

TABLE VI: Maximum Activity Concentration and Respective Distance to the Stacks.

Radionuclide	Maximum value (Bq/kg)	Station N°	Distance (km) and direction to the stacks	
Bi-212	49.81	24	06.0	SE
Bi-214	61.19	24	16.0	SE
Ac-228	72.84	24	16.0	SE
K-40	1895.07	24	16.0	SE
Pb-214	34.30	17	08.0	SE
Ra-224	54.23	17	08.0	SE
Tl-208	28.18	49	11.0	SE
Ra-226	69.70	44	06.5	NE
Bi-211	45.69	30	14.5	NE
Pb-212	46.91	8	03.0	NW

For all identified radionuclides the maximum values were detected at measurement stations 8, 17, 24 and 49. Measurement station 8 is located at 3 km to NW of the stacks while measurements stations 17, 24 and 49 are located at SE from the stacks.

The measurements stations 30 and 44, located at NE, showed maximum values for Bi-211 and Ra-226. However, this should be interpreted with caution as Bi-211 and Ra-226 were identified only in some of the total measurement stations (18 and 16 measurements, respectively). The same happens for Ra-224 that was only identified in 11 stations.

In general, radionuclides concentration is higher at NE and SE from the stacks, beyond the distance of 6 km from the stacks. A few hot spots occur within the 6 km area matching with the results obtained with the scintillation counter (SPP2).

Considering that the potential enhance in background radiation is related with the stacks emissions (fly ashes and PM₁₀) and consequently with the wind transport, dispersion and deposition, mainly from past situations, some considerations may be drawn.

The distance that the particles travel depends on meteorological conditions, particle size and stack height. The historical data referent to the study area indicates that the prevailing wind direction is from N-NW to S-SE with the exception for the month of October when the prevailing wind direction is from S-SW to N-NE. In autumn the prevailing wind blows from N with a frequency of 34.5%. Wind velocity is relatively constant during all year with long calm periods; stronger winds blow from N-NW and SW with maximum velocities during the winter season. The gamma radiation surveys were carried out in the beginning of autumn of 2011 and in the beginning of spring of 2012; the prevailing wind direction for these periods were from N-NW and from N-NE, respectively.

In atmospheric emissions the influence of stack height in the dispersion and deposition patterns has shown that about 25% of emitted particles from a 40 m stack fell within 5 km and 34% within 10 km.

Only 6% of the emitted particles from a coal power plant with a 300 m stack fell within the distance of 5 km [9]. 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 [9]. The height of the stacks (225 m) combined with wind patterns (which occurs predominantly from NW to SE) are expected to have transported the emissions up to long distances away from the stacks. In this case, the peak gamma radiation values detected near the stacks could not fall from the plume vertically. This anomaly is probably directly related with the coal deposit near the stacks (likely to be predominately from local fugitive sources in the form of windblown ash and dust).

High activity concentrations measured at NE, E and SE for distances up to 20 km from the stacks was influenced by the wind parameters as stacks height combined with wind patterns are expected to transport the emissions to long distances from the source. In fact, measurements results show that the detected concentration is higher for the stations located in the prevalent wind direction (SE) and for distances up to 20 km distant from the stacks.

CONCLUSIONS

The registered gamma radiation rate, at the 50 monitoring stations, vary from 20 to 98.33 c.p.s. and this corresponds to an exposure dose rate between 87.70 and 431.19 nGy/h with an average value of 230.08 nGy/h for the first survey and 228.22 nGy/h for the second one. These values are about 4 times higher than the world average rate (55 nGy/h), 3 times higher than the Portuguese average rate (84 nGy/h) and 6 times higher than the average rate for the Sines region (39 nGy/h). Additionally, the measured values are about 2 times higher than the established background (109.62 nGy/h).

The highest values for gamma radiation were registered in two stations located within the 6 km area at NW and at E from the stacks (stations 8 and 10, respectively) and for several stations within the 6-20 km area at SE from the stacks. The lowest values were detected at several stations placed at NW from the stacks in both survey areas.

Gamma spectrometry identified natural emitting nuclides as well as their decay products: Bi-211, Bi-214 Pb-214 Ra-226, Tl-208, Bi-212, Pb-212, Ra-224, Ac-228 and K-40. The activity concentration, at the 50 stations, varied from 0.93 to 73.68 Bq/kg for Ra-226 and Th-232 radionuclides series and from 84.17 to 1895.07 Bq/kg for K-40. The highest values measured within the 6 km area correspond to the same hotspots detected with the scintillometer counter but most of the highest values were measured for several stations within the 6-20 km area at SE from the stacks. The lowest values were measured at stations placed at NW from the stacks.

The results from both techniques used to measure the environmental and background radiation are in good agreement and show the same spatial pattern for gamma exposure rate and radionuclides activity concentration: the values are higher for the stations located in the prevalent wind direction (SE) and for distances up to 20 km distant from the stacks. The iso-concentration maps show precisely the existence of a gradient concentration beyond the 6 km distance from the stacks, up to 20 km.

In general, wind direction promotes the emissions dispersion in to the Atlantic Ocean. However, for higher wind speeds an inversion of wind direction occurs and the emissions are transported in the opposite direction from SW to NE which may explain the high values observed at NE and E as well. The stacks height and the dispersion by strong seasonal winds are responsible for the verified transport and dispersion of the fly ash up to distances of 20 km.

The only exceptions were verified for the two hotspots within the 6-km area and may be attributed to pontual sources such as the coal deposit near the stacks and the effluents discharged from the coal plant, mainly due to past situations. This should be subject to further research.

The results from seasonal sampling campaigns were compared, however, no seasonal variation could be observed and this requires a longer survey period in each season, for future works.

Sines coal-fired power has been a source of atmospheric emissions for many years. In the last five years these emissions decreased substantially. Nevertheless, past historical emissions are still present in the environment and must have contributed to the spatial dispersion pattern observed. Although recent measures mean that stacks height is no longer a significant point source of pollution, the contribution from other sources related to the coal combustion or to the coal plant as well as past historical emissions, may have lead to the accumulation in the environment increasing the natural background radiation.

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