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Assessment of the Radiological Impact on the Environment near a Storage Site of Coal Ashes in a Thermal Power Plant

Aziz Boukhair[1,2*], Laila Belahbib[1], Khadija Azkour[3], Hamid Nebdi[1], Mohammed Benjelloun[1], Abdelmjid Nourreddine[4]

[1]Laboratoire de Physique Nucléaire, Atomique et Moléculaire, Département de Physique, Faculté des Sciences Université Chouaïb Doukkali, El Jadida, Morocco
[2]Centre Régional des Métiers de l'Education et de la Formation, El Jadida, Morocco
[3]Ministère de l'Intérieur, Secrétariat Général, Province d'El Jadida, Morocco
[4]Groupe RaMsEs, Institut Pluridisciplinaire Hubert Curien (IPHC), Université de Strasbourg, Strasbourg, France
Email: *mouad.boukhair@gmail.com

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Abstract

The radiological impact of coal ashes, with enhanced natural radioactivity in the storage site, is due to the presence of naturally occurring radionuclides. Some of these radionuclides have a radioactive period of several million years and will, therefore, have time to migrate to the soil, atmospheric air, surface water, and groundwater. This impact depends mainly on the activity of these coal ashes, the duration of exposure to such waste, transfers to the air, and the leaching phenomenon by rainwater. In this study, and so as to assess the radiological impact of coal ashes of the storage site of the JLEC-Morocco thermal power plant on environment, some analyses are performed by alpha dosimetry and a digital dosimeter on samples of coal ashes, soil, atmospheric air, surface water and groundwater belonging to a perimeter of 10 km around that site. The obtained results show that, within the studied area, the radiological impact on the soil of the coal ashes of the storage site is insignificant even though the concentrations of radon in the near vicinity (1 to 2 km) are moderately important, and remain below 200 Bq/m^3. In the atmospheric air, this impact remains medium for the neighborhoods of the storage site (2 to 3 km) with radon activities superior to 10 Bq/m^3. These results also show that there may be a water contamination of wells located at the storage site without any transfer of radioactivity into the groundwater of the area studied where the concentrations of radon are less than 11.1 Bq/l.

Keywords

Storage Site, Coal Ashes, Environment, Radiological Impact, Soil, Water, Atmospheric

Air, LR115, Radon, Volumetric Activity, Exhalation of Radon

1. Introduction

In recent years, as far as the environment is concerned, there has been a growing awareness of professional and public risk of the radiological impact of non-nuclear industries which use, for various applications, raw materials naturally rich in uranium, thorium, or radium [1]. The industrial processing of these raw materials can generate large amounts of waste whose activity concentration can be higher than that of the original materials. This is called Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM).

In this study, we are interested in the industry dedicated to the production of electricity in power plants based on combustion coal that contains radionuclides with an activity of uranium and thorium between 5 and 300 $Bq{\cdot}kg^{-1}$ [2]. This production generates large amounts of solid residues including fly and bottom ashes constituting the majority of these wastes. Considering the increasing global demand for electricity, particularly in Morocco, the production of coal ashes is continuously increasing. The management of said production presents a major challenge for both industrialized and developing nations. Hence, it is highly recommended to store them in secure landfills. However, this recommendation raises two issues: while the first one has an economic character and is related to the management and development of landfills, the second issue is environmental and is related to the risk of contamination of the soil, atmospheric air, and surrounding waters due to the infiltration of other radioactive elements, such as radium, which exists in relatively large amounts in these coal ashes [3]-[5]. By radioactive decay, this radionuclide generates a radioactive gas: radon. It is now considered the main source of any population's exposure to natural radiation. Indeed, the 2006 UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) report suggests that, at an international level, radon accounts for about 52% of the overall average exposure to natural radiation [6]. On one hand, the radiological impact of the isotope 222 (48%) is significantly higher than that of the isotope 220 (4%). On the other hand, the isotope 219 is considered negligible. While disintegrating, radon emits alpha particles and produces solid radioactive descendants. These descendants can be attached to atmospheric aerosols and can be inhaled; thus, causing adverse health effects.

For this reason, in order to assess the radiological impact of the storage site of the coal ashes in the Jorf Lasfar Energy Company power plant (JLEC-Morocco) on the environment, our work will focus mainly on determining the volumetric activity and exhalation rate in terms of area and in terms of mass for ^{222}Rn in samples of coal ashes, soil, surface water and groundwater, and finally into the atmospheric air.

2. Materials and Methods

2.1. Presentation of the Storage Site of the Coal Ashes

The site of storage of the coal ashes in which we are interested is that of the thermal

power plant JLEC-Morocco. This is an old limestone quarry of 140,000 m^2 of area and more than 20 meters deeper than the surrounding land. The development of this controlled site near the place of production has the advantage of avoiding the dispersion of coal ashes into the environment.

In JLEC thermal power plant, coal combustion generates large masses of solid waste that exceeds 640,000 tons per year. Of this solid waste, fly ash is estimated to be around 500,000 tons. As for the bottom ash, the production rate exceeds 50,000 tons per year [7]. When this plant started in 1994, all the solid residues were released into the Atlantic Ocean. Since 1997, all of the bottom ash, and a part of the fly ash, which was not valued by some national cement, are stored in this site. The storage of this mixture of coal ashes is done by lockers. Each locker has a bottom and sides equipped with a geomembrane and a leachate drainage system so as to minimize the impact of the ash on the soil and on the quality of groundwater. To this day, three lockers are already completely filled and covered by a layer of about one meter of topsoil thickness. The fourth locker is on its way of being filled.

2.2. Radiological Impact of the Storage Site

The radiological impact of coal ashes, with enhanced natural radioactivity in the storage site, is caused by the presence of naturally occurring radionuclides. Some of these radionuclides have a radioactive period of several million years, and will, therefore, have enough time to migrate to the soil, surface water, and groundwater. Transfers are also equally possible to atmospheric air in the shorter term. This migration can contaminate the environment and have a radiological impact on the population. This impact depends mainly upon the activity of the coal ashes, the length of exposure to these wastes, transfers to air (takeoffs, suspensions), and leaching phenomenon by rainwater. This phenomenon could lead to the mobilization of significant amounts of radionuclides as a result of the important volumes of coal ashes involved. These radionuclides will migrate with rainwater entering the stock of coal ashes to mix with groundwater and cause the gradual enlargement of the affected area. In the present work, before assessing the radiological impact of the JLEC thermal power plant's storage site of the coal ashes on the environment, we first:

- Measured the specific activity of the radionuclide ^{226}Ra by gamma spectrometry and estimated the surface and mass exhalation rates of radon by alpha dosimetry in samples of fly and bottom ashes before they were stored in the site. These samples were provided to us by the JLEC thermal power plant in the month of January 2016.
- Calculated the activity concentration and the surface and mass exhalation rates of radon by alpha dosimetry in coal ashes, but this time they were stored in the site from 1997 to the present. To do so, during the month of January 2016, we collected in each locker, at different depths, three samples of coal ashes. This has led us to establish the variation in these exhalation rates depending on the storage time.

Secondly, and so as to achieve a relevant analysis, a study area of a perimeter of10 km at the storage site was defined to evaluate and clearly define the impact of coal ashes on

the receiving environment (Figure 1). For this, we calculated the activity concentration and the surface and mass exhalation rates of radon by alpha dosimetry in samples collected during the month of January 2016:

- The surface soil covering the three filled lockers and the surrounding soil,
- Surface water (leachate) and groundwater on the site and surrounding groundwater (wells).

Finally, we measured the average activity concentrations of radon in atmospheric air using a digital dosimeter, one meter above the ground and on different stations belonging to the specified area. These measures began on January and lasted until the end of June 2016.

2.3. Calculation of Radon Volume Activity

In this paragraph, we first describe the method and the experimental device used to calculate the volumetric activity of radon in samples of coal ashes before and after their storage at the site, the soil and surface water (leachate), and the groundwater (wells) in the study area.

To reach this objective, several pieces of 2 × 2 cm^2 of Solid State Nuclear Track detectors (SSNTD) LR115 type 2 non strippable, Kodak brand of 12 µm thick were exposed in sealed cylindrical "cans" of 5.5 cm diameter and 9.5 cm height by 50 g of each solid sample and 50 cm^3 of water sample.

Before any assessment and for homogeneous samples, coal ashes and soil are dried in

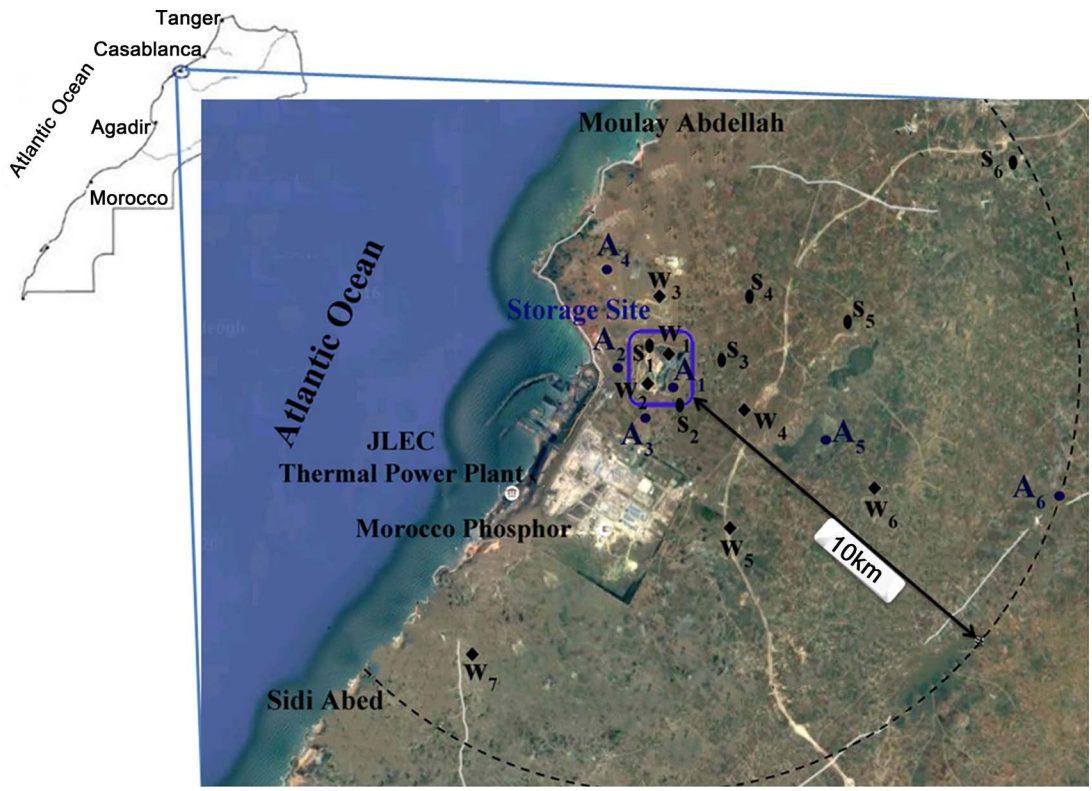

Figure 1. Delimitation of the study area at the level of the storage site of coal ashes.

stove at 40˚C during 24 hours, then crushed and sieved through a sieve of 100 µm. These samples, as well as the waters, are packaged in sealed radon containers for at least 4 weeks to establish secular equilibrium corresponding to seven half-lives of ^{222}Rn.

After two months of irradiation, the LR115 are chemically treated in a 2.5N sodium hydroxide solution during 100 min at a temperature of 60˚C. The reading of the developed films is performed using an optical microscope.

The density of traces per unit area and per unit time D_{LR} in the LR115, and the volumetric activity of radon A_V^{Rn} linked by the following relationship:

$$D_{LR} = \varepsilon_{LR}\left(\theta_c, E_\alpha\right) A_V^{Rn} \tag{1}$$

where $\varepsilon_{LR}\left(\theta_c, E_\alpha\right)$ is the efficiency of detection in a function of the critical angle of recording θ_c and the energy of the alpha particle E_α. This efficiency is equal to 0.0258 (traces·cm^{-2}·d^{-1})/(Bq·m^{-3}) [8].

Second, we used a the digital radon dosimeter CANARY [9] so as to measure the activity concentrations of radon in the atmospheric air. The principle of detection is based on diffusion of radon gas in a detection chamber. The alpha particles emitted by decay radon atoms, and which are of high energy, are detected by a silicon photodiode associated with a set of electronic modules for shaping, amplifying, and storing pulses. This dosimeter indicates the average value of radon activity concentration in Bq/m^3 on a daily, weekly, and on a full year basis. In our case, the average volume activities for a given area are measured for a period of one month at one meter from the ground.

2.4. Calculation of Radon Exhalation Rate

The exhalation rate in terms of area of ^{222}Rn (Bq·m^{-2}·h^{-1}) is determined by the following formula [10] [11]:

$$E_S = \frac{A_V^{Rn} V \lambda_{Rn}}{S_e\left[t + \left(\frac{1}{\lambda_{Rn}}\right)\left(e^{-\lambda_{Rn} t} - 1\right)\right]} \tag{2}$$

With A_V^{Rn} is the volumetric activity of radon (Bq·m^{-3}·h); V is the effective volume of can (m^3); λ_{Rn} is the decay constant for radon (h^{-1}); S_e is the sample surface (m^2) and t is the exposure time (h).

The exhalation rate in terms of mass of ^{222}Rn (Bq·kg^{-1}·h^{-1}) is determined by:

$$E_M = \frac{A_V^{Rn} V \lambda_{Rn}}{M\left[t + \left(\frac{1}{\lambda_{Rn}}\right)\left(e^{-\lambda_{Rn} t} - 1\right)\right]} \tag{3}$$

where M is the mass of the sample in kg.

3. Results and Discussion

The specific activities of radionuclide ^{226}Ra determined by gamma ray spectrometer using the Broad Energy Germanium detector (BEGe) of planar type in samples of fly and bottom ashes from the JLEC thermal power plant before storing them in the site, the

volume of activities and surface and mass exhalation rates of radon in these samples are summarized in Table 1.

It is noted that the activities of ^{226}Ra in samples of fly and bottom ashes are respectively of the order of 149 Bq/kg and 84 Bq/kg. These activities are well above the permissible activity which is of the order of 40 Bq/kg [12]. As to activity concentrations and the surface and mass exhalation rates of radon in samples of the bottom ash, which are respectively of the order of (465 Bq/m^3, 381 mBq·m^{-2}·h^{-1} and 18 mBq·kg^{-1}·h^{-1}), they are still higher than those of the samples of fly ash (337 Bq/m^3, 276 mBq·m^{-2}·h^{-1} and 13 mBq·kg^{-1}·h^{-1}). The specific activities of ^{226}Ra and the exhalation rate in terms of area and mass of radon measured in the samples of fly and bottom ashes from the JLEC thermal power plant are comparable to those found in other thermal power plants worldwide [3] [12] [13].

Having analyzed samples of coal ash before storage, we present in Table 2 the volumetric activity and exhalation rate in terms of area and mass of radon measured in coal ashes samples taken from three racks of storage site of different depths. We note from these values that the average radon activity concentration varies from 766 Bq/m^3 in locker 1 (corresponding to the storage period from 1997 to 2008) to 704 Bq/m^3 in locker 2 (2009-2011) and up to 541 Bq/m^3 in locker 3 (2012-2014). It is the same for the exhalation rate of radon. Indeed, the surface and mass exhalation rates measured in the samples of coal ash in the three storage lockers vary respectively from (609 - 430 mBq·m^{-2}·h^{-1}) to (29 - 20 mBq·kg^{-1}·h^{-1}). These differences in volume activity and radon

Table 1. Specific activities of ^{226}Ra (A_c^{Ra}), volume activity (A_V^{Rn}) and the exhalation rate in terms of area (E_S) and mass (E_M) of radon in fly ash (FA) and bottom ash (BA) before storage compared to other works.

Thermal Power Plants	Samples	A_c^{Ra} (Bq/kg)	A_V^{Rn} (Bq/m^3)	E_S (mBq·m^{-2}·h^{-1})	E_M (mBq·kg^{-1}·h^{-1})
JLEC-Morocco[3]	FA	149 ±26	337 ± 27	276 ± 22	13 ± 1
	BA	84 ± 16	465 ± 38	381 ± 31	18 ± 2
India (2015) [13]	FA	14.05 to 26.12			23.37 to 42.73
India (2013) [14]	FA	118.6 ± 7.4	431.7 ± 35.5	155.5 ± 12.8	6.0 ± 0.5
India (2010) [15]	FA	99 to 203	214 to 590	138 to 381	7.8 to 21.6
Spain (2009) [4]	FA	191 ± 9			
	BA	149 ± 6			
Turkey (2008) [16]	FA	149 ± 2			
	BA	50 ± 1			
China (2006) [17]	FA	112.2			
	BA	93.4			
Greece(2004) [18]	FA	261			14.4
	BA	114			82.8
India (2002) [19]	FA		748 to 1377	483 to 914	27 to 52

Table 2. Volumetric activity and the exhalation rate of radon in coal ashes samples from the three storage lockers.

Rack Number	Storage period	Removal level	A_V^{Rn} (Bq/m^3)	E_S (mBq·m^{-2}·h^{-1})	E_M (mBq·kg^{-1}·h^{-1})
1	1997-2008	Top	553 ± 45	440 ± 35	21 ± 2
		Middle	625 ± 51	496 ± 40	24 ± 2
		Bottom	1121± 94	891 ± 71	42 ± 3
		Average value	766 ± 64	609 ± 50	29 ± 2
2	2009-2011	Top	621 ± 51	494 ± 39	23 ± 2
		Middle	643 ± 53	511 ± 42	24 ± 2
		Bottom	850 ± 70	675 ± 54	32 ± 3
		Average value	704 ± 58	560 ± 46	27 ± 2
3	2012-2014	Top	443 ± 36	352 ± 28	17 ± 1
		Middle	468 ± 38	372 ± 30	18 ± 1
		Bottom	714 ± 59	567 ± 45	27 ± 2
		Average value	541 ± 44	430 ± 35	20 ± 2

exhalation rate may be due to the type of coal used in the combustion by the JLEC power plant [20]. There was also a high concentration of radon measured in samples collected at the bottom of the three lockers, consequently, high rates of surface and mass exhalation of radon, values vary respectively for the volume concentration and the surface and mass exhalation rates of radon (1121 - 714 Bq/m^3), (891 - 567 mBq·m^{-2}·h^{-1}), and (42 - 27 mBq·kg^{-1}·h^{-1}).

The volumetric activity and the exhalation rate of radon measured in surface water (leachate), groundwater (wells) and soil samples belonging to the storage site and the area studied are gathered in **Table 3**.

There is a high volume concentration of radon in the leachate with an activity of about 4683 Bq/m^3 and, therefore, high rates of surface and mass radon exhalation, respectively of the order of 3147 mBq·m^{-2}·h^{-1} and 189.36 mBq·kg^{-1}·h^{-1}. This concentration is mainly caused by the enormous volume of coal ash stored since 1997, the content and mobility ^{226}Ra filling the lockers by rainwater, and the disposal of the latter by the drainage network to the leachate pond.

The values and volumetric activity and the exhalation rate in terms of area and mass of radon in the wells in the studied area are very low (61 to 157 Bq/m^3, 39 to 101 mBq·m^{-2}·h^{-1} and 2.36 to 6.08 mBq·kg^{-1}·h^{-1}) compared with those registered at the well located at the storage site (1684 Bq/m^3, 1132 mBq·m^{-2}·h^{-1} and 68.09 mBq·kg^{-1}·h^{-1}). The result is that currently there is no transfer of radioactivity into the groundwater of the studied area, while there may be water contamination of wells located at the storage site. Note that the values of activity concentrations of water in the area studied are lower than the permissible limit (11.1 Bq/l) [21].

There are also strong variations of the activity concentration and the surface and mass exhalation rates of radon in soil samples taken from the soil of the storage site and

its immediate environment, varying from (262 to 32 Bq/m^3), (223 to 26 mBq·m^{-2}·h^{-1}) and (10.60 to 1.24 mBq·kg^{-1}·h^{-1}), respectively. It shows that even if these values are moderately important in close proximity of the storage site (1 to 2 km), they remain below the permissible limit 200 Bq/m^3 [22].

The average activity concentrations of radon in atmospheric air measured using the dosimeter digital CANARY in the various stations of the study area is summarized in Table 4.

We notice that there is a significant variation of the average radon activity concentration in the storage site (43 Bq/m^3) and its immediate environment (4 Bq/m^3) with a variation factor of 11. This variation may be due to the dispersion of the particles from coal ashes caused by the movement of construction equipment during the transport and storage of ashes particularly, in the vicinity of the storage site (2 to 3 km), and this could be feared, especially during windy periods. The average activity concentrations

Table 3. Volumetric activity and the exhalation rate in terms of area and mass of radon levels in samples of water and soil in the area studied.

Samples	Symbols	Locality	A_V^{Rn} (Bq/m^3)	E_S (mBq·m^{-2}·h^{-1})	E_M (mBq·kg^{-1}·h^{-1})
Water	W1	Leachate pond	4683 ± 374	3147 ± 251	189.36 ± 15.12
	W2	Well on site	1684 ± 137	1132 ± 92	68.09 ± 5.54
	W3	Well 1 km Site	74 ± 6	48 ± 4	2.87 ± 0.23
	W4	Well 2 km Site	157 ± 13	101 ± 8	6.08 ± 0.49
	W5	Well 4 km Site	105 ± 8	67 ± 5	4.05 ± 0.32
	W6	Well 7 km Site	87 ± 7	56 ± 5	3.38 ± 0.27
	W8	Well 8 km Site	61 ± 5	39 ± 3	2.36 ± 0.19
Soil	S1	On the site	262 ± 21	223 ± 18	10.60 ± 0.85
	S2	0.5 km Site	157 ± 13	128 ± 10	6.06 ± 0.49
	S3	1 km site	97 ± 8	79 ± 6	3.75 ± 0.30
	S4	2 km site	75 ± 6	61 ± 5	2.90 ± 0.24
	S5	4 km site	43 ± 3	35 ± 3	1.66 ± 0.13
	S6	10 km site	32 ± 3	26 ± 2	1.24 ± 0.10

Table 4. Average volumetric activity of ^{222}Rn in atmospheric air measured in the various stations of the study area.

Station	Symbols	Period	A_V^{Rn} (Bq/m^3)
On site	A1	January 2016	43 ± 4
0.5 km site	A2	February 2016	25 ± 3
1 km site	A3	March 2016	21 ± 2
2 km site	A4	April 2016	18 ± 2
4 km site	A5	May 2016	8 ± 1
10 km site	A6	June 2016	4 ± 1

measured in this area are greater than 10 Bq/m^3, the average activity value of radon in outdoor air at the Earth's surface [23]. Therefore, the radiological impact of coal ash storage site on the atmospheric air is set to medium.

4. Conclusions

When we started this work, our objective was the assessment of the radiological impact of the coal ash storage site at the JLEC-Morocco thermal power plant on the environment, by determining the activity concentrations and the rates of surface and mass exhalation of radon in samples of coal ash before and after storage in the site:

- The values obtained before storage for bottom ash are of the order of (465 Bq/m^{-3}, 381 mBq·m^{-2}·h^{-1} and 18 mBq·kg^{-1}·h^{-1}), respectively and remaining higher than those obtained for the fly ash (337 Bq/m^{-3}, 276 mBq·m^{-2}·h^{-1} and 13 mBq·kg^{-1}·h^{-1}).
- The values obtained in lockers for the storage site vary (766 Bq/m^3, 609 mBq·m^{-2}·h^{-1} and 29 mBq·kg^{-1}·h^{-1}), respectively in locker 1 corresponding for the period of storage in 1997-2008 to (541 Bq/m^3, 430 mBq·m^{-2}·h^{-1} and 20 mBq·kg^{-1}·h^{-1}) and in the locker 3 corresponding for the period 2012-2014. This variation may be due to the nature of the coal used in the combustion by the JLEC thermal power plant.

In order to assess the radiological impact of the coal ash on the environment, we have used alpha dosimetry and by the CANARY digital dosimetry so as to measure the concentrations activities and surface and mass exhalation rates of radon in samples of soil, surface water and groundwater and the atmospheric air belonging to an area of 10 km at the level of the storage site. We notice:

- A strong variation of the activity concentration and surface and mass exhalation rates of radon in soil samples taken from the soil of the site and its immediate environment, a changing values from (262 to 32 Bq/m^3), (223 to 26 mBq·m^{-2}·h^{-1}), and (10.60 to 1.24 mBq·kg^{-1}·h^{-1}), respectively.
- A high volume concentration and, therefore, high rates of surface and mass exhalation of radon in the leachate and in the well located in the storage site. The values are of the order of (4683 Bq/m^3, 3147 mBq·m^{-2}·h^{-1}, 189.36 mBq·kg^{-1}·h^{-1}) and (1684 Bq/m^3, 1132 mBq·m^{-2}·h^{-1}, 68.09 mBq·kg^{-1}·h^{-1}), respectively. This concentration is mainly caused by the enormous volume of coal ash stored since 1997 and the content and mobility of ^{226}Ra filling lockers by rainwater.
- A significant variation in the average radon activity concentration in the atmospheric air on the storage site (43 Bq/m^3) and its immediate environment (4 Bq/m^3). This variation may be due to the dispersion of the particles from coal ashes, especially in the vicinity of the storage site (2 to 3 km).

Based on these factors, we deduce that the radiological impact of the storage site of coal ash on the studied perimeter:

- Is insignificant for the same soil where concentrations of radon in close proximity of the storage site (1 to 2 km) are moderately important, but remain below 200 Bq/m^3,
- Remains medium in atmospheric air and especially in the vicinity of the storage site (2 to 3 km) with radon concentrations above 10 Bq/m^3,

- Shows perhaps a contamination of water wells located at the storage site, while stressing that there is no transfer of radioactivity into the groundwater of the area studied where the concentrations of radon are less than 11.1 Bq/l.

It follows from this study that it would be important to install radiation monitoring in the long term for the surrounding environment of the storage site so as to detect any abnormal conditions or developments.

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