doi.org/10.15392/2319-0612.2024.2482
2024, 12(3) | 01-14 | e2482
Submitted: 2024-04-29
Accepted: 2024-09-23

Decommissioning and demolition of a dicalcium phosphate plant: aspects of radiological protection and safety

Trindade, R. [a*]; Narciso, J. [a]; Lopes, D. [a]; Marques, S [a]; Albuquerque, C [a]; Schiappa, J [a]; Matos, A [a]; Moreira, J [a].

[a] Ambimed – Gestão Ambiental, Lda., 2560-587 Torres Vedras, Portugal

*Correspondence: romao.trindade@stericycle.com

Abstract: The detection of ionising radiation in materials from the decommissioning of a dicalcium phosphate plant meant that this whole operation had to be overhauled in terms of radiological protection and safety. Thus, an operations plan was drawn up that included: initial training for all workers involved, measurements of dose rates and a set of analyses by gamma and alpha spectrometry. Samples of "dry sludge" and debris were analysed for the identification and quantification of the radionuclides present, samples of workers' urine were analysed for the detection of any internal contamination, samples of washing water and air filters were analysed for the detection and control of any radioactive atmospheric contamination. In addition, individual dosimeters were distributed to workers. The extent of contamination was identified and the areas where higher values were detected were properly marked. From the results obtained (radiation dose rates and activity concentrations), it was possible to establish categories and separate the contaminated materials according to their physical characteristics. After the plant decommissioning, the resulting waste with dose rates higher than the effective dose limits for the public was packed in *big bags*. These bags were divided in 3 groups according to the external radiation dose rates at contact. This segregation allowed the competent authority to release a significant number of *big bags* from regulatory control. The remaining *big bags* are stored in two metal containers subject to regular radiological monitoring. The objective of this work is the presentation of a factual article where the methodology implemented to deal with this situation, which had never occurred in Portugal, is described, having only the Portuguese legislation as a reference.

Keywords: decommissioning, radiation protection, dicalcium phosphate, radiation monitoring.

doi.org/10.15392/2319-0612.2024.2482
2024, 12(3) | 01-14 | e2482
Submitted: 2024-04-29
Accepted: 2024-09-23

Desmantelamento e demolição de uma fábrica de fosfato dicálcico: aspetos de proteção e segurança radiológica

Resumo: A deteção de radiação ionizante em materiais provenientes do desmantelamento de uma fábrica de fosfato dicálcico fez com que toda esta operação tivesse de ser reformulada em termos de proteção e segurança radiológica. Elaborou-se um plano de operações que incluiu: ação de formação inicial a todos os trabalhadores envolvidos, medições de débitos de dose e um conjunto de análises por espectrometria gama e alfa. Foram analisadas amostras de "lamas secas" e entulhos para identificação e quantificação dos radionuclídeos presentes, de urinas dos trabalhadores para deteção de qualquer contaminação interna, de águas de lavagem e filtros de ar para deteção e controlo de qualquer contaminação atmosférica radioativa. Foram distribuídos dosímetros individuais aos trabalhadores. A extensão da contaminação foi identificada e as áreas onde se detetaram valores mais elevados foram devidamente sinalizadas. A partir dos resultados obtidos (débitos de dose de radiação e concentrações de atividade) foi possível estabelecer categorias e separar os materiais contaminados de acordo com as suas características físicas. Após o desmantelamento da instalação, os resíduos resultantes que apresentavam débitos de dose superiores aos limites de dose efetiva para o público foram acondicionados em *big bags*. Estes sacos foram divididos em 3 grupos, de acordo com os débitos de dose de radiação externa ao contacto. Esta separação permitiu que a autoridade competente pudesse liberar do controlo regulamentar um número significativo de *big bags*. Os restantes encontram-se acondicionados em dois contentores metálicos sujeitos a monitorização radiológica regular. O objetivo deste trabalho é a apresentação de um artigo factual onde é descrita a metodologia implementada para lidar com esta situação, que nunca tinha acontecido em Portugal, tendo apenas como referência a legislação portuguesa.

Palavras-chave: desmantelamento, proteção radiológica, fosfato dicálcico, monitorização radiológica.

1. INTRODUCTION

The detection of ionising radiation, in the gantry of a steel industry, in materials from the decommissioning and demolition of a dicalcium phosphate plant, essential in the animal feed industry, meant that the entire initially planned operation was subject to special care and had to be reformulated in terms of radiological protection and safety.

In the on-site monitoring, it was verified that the radiation originated from the radioactivity existing in a kind of light-coloured "dry sludge" deposited over the years on the walls of the reactors, which are integral parts of the phosphate production chain, and which was dispersed.

The radionuclide ^{226}Ra has a half-life period of 1600 years, it is essentially an alpha emitter but it is also a gamma emitter. This radionuclide is considered NORM *(Naturally Occurring Radioactive Material)*, the designation given to radioactive elements of natural origin. The concentration of NORM in the waste, by chemical processes, transforms it into TENORM *(Technologically Enhanced Naturally Occurring Radioactive Materials)*.

The objective of this work is the presentation of a factual article where the methodology implemented to deal with this situation, which had never occurred in Portugal, is described, having only the Portuguese legislation as a reference.

2. MATERIALS AND METHODS

In a first radiological evaluation, using portable detection equipment, gamma radiation dose rate values were higher than the dose limits for members of the public defined by the Portuguese Environment Agency (0,5 μSv/h). With a portable NaI detector, the radionuclide ^{226}Ra was identified as the main radiation emitter.

The extension of contamination was identified and the areas where highest radiation levels were detected were properly marked.

The unreleased debris, sludge and other material were packed in big bags stored in two containers of 40' (12 m) and 30' (9 m) in length.

All materials resulting from the demolition of the factory (Figure 1) were packed in duly numbered *big bags*. Around each *big bag*, a set of points was defined to measure the dose rates of ionizing radiation upon contact, with 10 values being recorded for each of these points. The measurements were made on contact because the objective was to relate doses with activities in order to apply the exemption and clearance criteria defined by the Portuguese Environment Agency. After recording the values, the maximum and minimum values were noted, and the arithmetic mean of the dose rates was calculated, as well as the respective standard deviation.

The *big bags* were divided into 3 groups according to the dose rates recorded:

- dose rate is less than 1,25 μSv/h;
- dose rate between 1,25 and 2,5 μSv/h;
- dose rate of more than 2,5 μSv/h.

From each of the groups of *big bags*, samples were taken to determine the activity concentration, by gamma spectrometry, in order to establish a linear relationship with the dose rate. Since the samples were very heterogeneous, the stirs were mixed with a shovel so that the contents of the *big bags* were as homogeneous as possible. For workers' washing water, the same spectrometric analysis methodology was adopted.

On the first day of the intervention, Panasonic UD-802A individual thermoluminescent dosimeters were distributed to all workers involved. The dosimeters were read at the end of the work.

At the beginning and end of the work, urine was collected from workers involved in the entire process, which were subsequently analysed by alpha and gamma spectrometry, in order to establish a baseline or reference on the presence of radioactivity and possible internal contamination of workers. In order to prevent internal contamination, workers wore personal protective equipment such as gloves, suits, boots and dust masks with forced breathing, which covered the entire face, including the eyes.

Figure 1: Dried sludge in reactor (A) and debris (B) for composite sample

Regarding the different types of debris (concrete and brick), composite samples were collected at various points of the work and analysed. PVC samples were also taken from the reactors for spectrometric analysis.

For the dust collection, two *Sequential High Volume Collector CAV-AMSB Mass Flow Control* air samplers were used. These samplers were identified as "Work" and "Public", as one was located on the land where the demolition was taking place and the other in a public place (4 m above ground). The samplers operated 24 hours a day with a flow rate of 30 m^3/h. The air filters were also analysed by gamma spectrometry.

All spectrometry analyses were performed by accredited laboratories.

The following equipment was used to measure the radiation dose rates and to initially identify the radionuclides present, respectively:

- Geiger Muller Detector Atomtex AT6130C;
- NaI Detector CANBERRA InSpector 1000.

3. RESULTS AND DISCUSSIONS

3.1. Dosimetry

The tests of the individual thermoluminescent dosimeters were carried out by the dosimetry laboratory of Ambimed – Gestão Ambiental, Lda. The results are presented in Table 1.

Table 1 : Results of individual dosimetry of workers.

Worker	Type of dosimeter	Equivalent dose	Total Dose [mSv]
I	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
II	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
III	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
IV	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
V	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
VI	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10
VII	Whole Body	Hp (0,07)	<0,10
		Hp (10)	<0,10

The results are indicated as <0,10 mSv as it is the minimum detection limit of the dosimeter.

After analysing the results, we can affirm that there was no irradiation of the workers with functions in the demolition of the facility.

3.2. Urine analysis

The workers involved were subjected to urine analysis by alpha and gamma spectrometry. Samples were collected at the beginning and end of the work.

3.2.1. Alpha spectrometry

The detection assays of ^{226}Ra by alpha spectrometry were carried out at the Laboratory of Environmental Radioactivity of the Universidad de Extremadura, in Cáceres (Spain). Table 2 shows the results of the urine analysis by alpha spectrometry collected at the beginning and at the end of the work.

Table 2 : Results of alpha spectrometry in the urine of workers..

Sample	^{226}**Ra**		
	Specific Activity [Bq/l]		**Minimum Detectable Activity, MDA [Bq/l]**
	Start	**End**	
Worker I	< 0,012	< 0,012	0,012
Worker II	< 0,011	< 0,011	0,011
Worker III	< 0,012	< 0,012	0,012
Worker IV	< 0,007	< 0,007	0,007
Worker V	0,016 ± 0,007	0,016 ± 0,007	0,006
Worker VI	< 0,015	< 0,015	0,015
Worker VII	< 0,010	< 0,010	0,010

3.2.2. Gamma spectrometry

The detection assays of ^{226}Ra, ^{228}Ra and ^{235}U by gamma spectrometry were carried out at the Laboratory of Radiological Protection and Safety of Instituto Superior Técnico, in Loures. Table 3 shows the results of the urine analysis by gamma spectrometry, collected at the beginning and at the end of the work. The work lasted 57 days.

Table 3 : Results of gamma spectrometry in the urine of workers.

Sample	Specific Activity [Bq/kg]					
	^{226}Ra MDA : 1,4		^{228}Ra MDA : 2,0		^{238}U MDA : 1,9	
	Start	End	Start	End	Start	End
Worker I	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker II	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker III	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker IV	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker V	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker VI	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9
Worker VII	< 1,4	< 1,4	< 2,0	< 2,0	< 1,9	< 1,9

The results of the urine analysis of the workers demonstrate that there was no internal contamination of the workers with functions in the demolition of the factory.

3.3. Analysis of air filters

The dust deposited in the filters removed from the "Work" site and from a "Public" site was analysed by gamma spectrometry at the Radiological Protection and Safety Laboratory of the Instituto Superior Técnico, in Loures. The results are presented on Table 4.

Table 4 : Results of gamma spectrometry in air filters.

Sample	Specific Activity [Bq/filter]					
	^{7}Be	^{137}Cs	^{210}Pb	^{226}Ra	^{228}Ra	^{238}U
	MDA : 1,4	**MDA : 0,035**	**MDA : 2,7**	**MDA : 0,12**	**MDA : 0,067**	**MDA : 0,21**
Filter 1 « Work »	12,7	< 0,035	< 2,7	< 0,12	< 0,067	< 0,21
Filter 2 « Work »	23,3	< 0,035	4,8	0,64	< 0,067	< 0,21
Filter 3 « Work »	7,2	< 0,035	< 2,7	1,4	< 0,067	< 0,21
Filter 1 « Public »	22,0	< 0,035	< 2,7	0,181	< 0,067	< 0,21
Filter 2 « Public »	10,6	< 0,035	< 2,7	< 0,12	< 0,067	< 0,21

The results of the analysis of the air filters demonstrate that there was no air contamination resulting from the demolition of the facility.

3.4. Washing water

The washing water of the workers and tools was collected in containers and analysed by gamma spectrometry for detection of possible radioactive contamination at the Laboratory of Radiological Protection and Safety of Instituto Superior Técnico. Table 5 shows the results obtained.

Table 5 : Results of gamma spectrometry in washing water.

Sample	Specific Activity [Bq/Kg]			
	^{210}Pb	^{226}Ra	^{228}Ra	^{238}U
	MDA : 71	**MDA : 1,6**	**MDA : 2,0**	**MDA : 2,9**
Water 1	< 71	< 1,6	< 2,0	< 2,9
Water 2	90	< 1,6	< 2,0	< 2,9
Water 3	< 71	< 1,6	< 2,0	< 2,9

After the analysis, all the water was discarded as normal waste.

3.5. *Big Bags*

The samples of *big bags* were analysed by gamma spectrometry to quantify the concentration of the radionuclides present, further characterization and eventual classification as radioactive waste.

These residues were packed in *big bags* and, before any spectrometry analysis was carried out, all *big bags* were monitored in order to determine the external radiation dose rates and estimate concentrations for each *big bag*.

The mean value of the dose rates results from the arithmetic mean of a set of 10 measurements around each *big bag*. As can be easily seen, the concentration values and dose rates are very variable, some of them far exceeding the dose limit for exposed workers defined by the Portuguese Environment Agency (10 µSv/h).

The values of the dose rates were divided into three intervals and the *big bags* were grouped according to these intervals.

Thus, we have:

- 42 *big bags* whose dose rate is less than 1,25 µSv/h;

- 16 *big bags* in the range between 1,25 and 2,5 μSv/h;
- 45 *big bags* with a dose rate of more than 2,5 μSv/h.

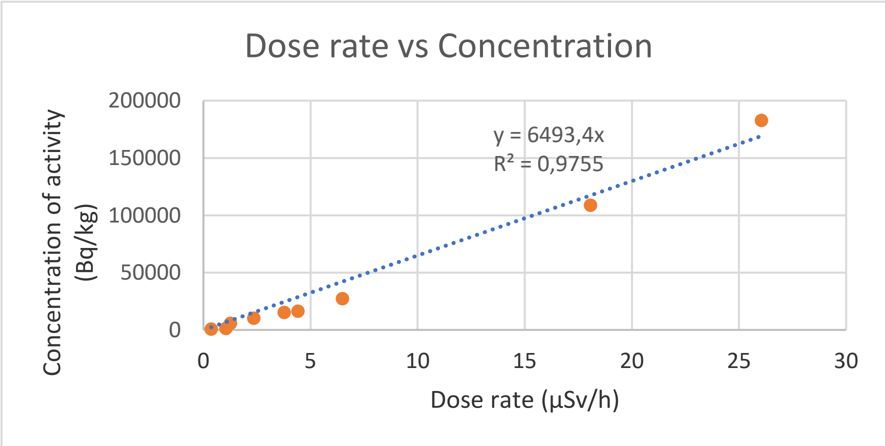
From these values, *big bags* were chosen from which the samples were taken to be analysed at the Laboratory of Radiological Protection and Safety of Instituto Superior Técnico. Table 6 shows the results of the spectrometric analysis for the radionuclide ^{226}Ra and the mean values of the dose rates measured with portable equipment.

Table 6 : Results of the spectrometric analysis of *big bags* for the radionuclide ^{226}Ra .

Sample	Activity ^{226}Ra [Bq/kg]	Dose Rate [μSv/h]
Big bag 63	807	0,36
Big bag 35	10428	12,83
Big bag 55	50675	21,85
Big bag 34	182747	26,04
Big bag 32	108958	18,06
Big bag 24	15309	3,77
Big bag 22	16373	4,41
Big bag 14	10320	2,35
Big bag 11	27239	6,48
Big bag 44	1297	1,04
Big bag 2	50290	30,00

Figure 2 relates the dose rate values measured with portable equipment and the activity concentration in previously chosen *big bags*, calculated by gamma spectrometry in an accredited laboratory.

Figure 2: Correlation between dose rates and activity concentration of the *big bags*.



From the equation $y = 6493{,}4\ x$, where x is the mean dose rate in the *big bag* and y is its activity concentration, the activity of all the *big bags* in the work was estimated.

Figure 3: *Big Bags*. packed in a metal container.

4. CONCLUSIONS

During the demolition of the dicalcium phosphate plant, it was necessary to carry out several radiological monitoring activities, since radiation dose rate values higher than the dose limit, for the public and for exposed workers, were detected.

Several analyses were carried out for radiological control of the tasks, in order to ensure the protection and radiological safety of workers and the environment.

The results of these analyses showed that there was no external or internal contamination of workers and the environment. The results of individual dosimetry also did not reveal any significant irradiation of the workers involved.

The various rubble and "dry sludge" were properly separated, and some of them were packed in *big bags* so that the regulator, the Portuguese Environment Agency, could classify them as radioactive waste or not.

Of the 103 existing *big bags*, some 41 could be excluded from regulatory control using the exemption and release criteria. The rest were packed in 2 metal containers of 30" (9 m) and 40" (12 m). These *big bags* are awaiting a final decision on their storage.

The results allow us to conclude that the methodology implemented was adequate since there was no internal or external contamination of the workers and the environment. Since there had never been a case like this in Portugal, this work becomes truly important as it can serve as a reference for cases that may happen in the future.

REFERENCES

[1] IAEA-TECDOC-1712, Management of Norm Residues, Vienna, June 2023, ISBN 978-92-0-142710-6.

[2] Decree-Law No. 156/2013, of November 5: establishes the legal and regulatory framework for the responsible and safe management of spent fuel and radioactive waste and transposes into Portuguese law the provisions of Council Directive 2011/70/Euratom of 19 July 2011.

[3] Ordinance No. 138/2019, of May 10: approves the exemption and release criteria.

[4] Decree-Law No. 108/2018, of December 3: establishes the legal framework for radiological protection, as well as the powers of the competent authority and the inspection authority for radiological protection, transposing into Portuguese law Council Directive 2013/59/Euratom of 5th December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation.

[5] Decree-Law No. 81/2022, of December 6: amends the legal regime Portuguese of radiological protection, adapting the rules on incompatibilities to the administrative offence regime and application.

[6] Resolution of the Council of Ministers No. 129/2022, of December 20: approves the National Portuguese Program for the Management of Spent Fuel and Radioactive Waste.