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THE CHARACTERIZATION OF THE RADIOACTIVITY IN THE CACICA SALT MINE

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Abstract. This paper presents the radioactive characterization (alfa-beta, gamma, radon and tritium) of the Cacica salt mine located in the Suceava county for speleotherapeutycal and medical purposes and for balneary and climatic tourism. The measurements have been made in situ and on samples of salt and water taken from the interior of the mine. These measurements were made as a part of a complex study from a radioactive, medical and biological point of view for an inovative use of the factors found in salt mines and caves that have a therapeutical potential in healthcare and balneoclimateric tourism.

Key words: radiological characterization, dosimetric systems, radiological and dosimetric measurements.

1. INTRODUCTION

The results presented in the paper show the radioactive measurements of the natural radiation background from the Cacica salt mine, Suceava County, Romania for speleotherapeutycal and medical purposes and for balneary and climatic tourism. These results includ radioactive measurements: alfa-beta, gamma, radon and tritium which were made both in situ and in water and rough salt samples. The following equipment and speciliased measurement systems was used for measurements both in the salt mine and in the loboratory: universal monitor *Umo LB 123, Berthold Technologies*, alpha measuring system, global low background beta beta measuring system *ORTEC PROTEAN MPC-2000-DP*; spectrometric gamma system with GeHP detector for the measurement of the activity of gamma

issuing radionuclide found in the samples, *ORTEC* model; portable monitoring system for the measurement of atmospheric radon *PYLON AB-5, Pylon Electronics Inc*; air dehumidifier *TROTEC*, *TTK 100S, TROTEC GmbH & Co. KG* [1].

The measurements made took into account the local variation of the distribution of the radionuclide and were averaged, the obtained result being closer to the specific area of the chosen environment.

The Cacica salt mine in which the radiological measurements were made is characterised by:

– temperature: between 10.2 ^{0}C and 11.5 ^{0}C;
– pressure: between 1008 and 1009.6 hPa;
– humidity: between 74% - 79%;
– the presence of saline aerosols;
– lighting: artificial;
– own ventilation system.

The work and calibration procedures were conducted in conformity with the work procedures of the *SALMROM* laboratory [1,2].

2. METHOD AND EXPERIMENTAL DATA

The measurements made consisted in: measurements of the armospherical Radon concentration, gamma spectrometric measurements with HPGe detector, Alpha-beta global measurements, measurements of the radiation fond, and tritium measurements.

2.1. THE MEASUREMENT OF THE CONCENTRATION OF THE ATMOSPHERICALLY RADON

The measurements of volume activity of atmosferical radon were made with the *Pylon AB 5* portable system with the help of the *CPRD* device in several areas from the interior of the salt mine and the measuring points, their coordinates and the environment conditions are presented in Table 1.

After the execution of the measurements and the transfer of the data on a laptop (relation interface) with the help of specialized software *Transfer Utility* 1.1 (DTU), Windows *Hyperterminal* or *TelNet* for *Windows Vista*, the *EXCEL* file *run#.(cod/no.)* has been used to present the results of the table measurements and the calculated errors/deviations.

Table 2 shows the measuring locatons with the measured values of the radon concentrations and with the calculatee errors. Thus we calculated: $S(n–1)$ – experimental standard deviation, $S(n–1)$ [%] – relative experimental standard

deviation, *S*(med) – experimental standard deviation of the mean, *S*(med) [%] – relative experimental standard deviation; *S*(Poisson) – Poisson relative error, global absolute error, etc., which are presented for each measuring (Table 2). *S*(Poisson) is given for comparison with the value of the standard deviation on the average value, for the purpose of statistical assessment of the average values versus the total number taken.

In Tables 3–8 (Figs. 1–6), there presented the diurnal variations of the Rn concentrations underground in the chosen measuring points on 20 minutes time intervals. It can be observed that when the ventilation is on in the salt mine, the concentration of Radon drops significantly, and when it is off it rises. The data recorded in the first 3 hours were not taken into account because this time is necessary for the system to reach its equilibrium point [4].

Table 1

Measuring points with coordinates and environment conditions

No.	MEASURING POINT/ LOCATION	COORDINATES	DEPTH [m]	AVERAGE TEMPERATURE [°C]	AVERAGE PRESSURE [kPa]	AVERAGE HUMIDITY [%]	AVERAGE WIND SPEED [m/s]
1	Salt lake (swimming pool) Point #1	47°38'06.51" N 25°53'52.55" E	32	10.2	1008	79	0.8
2	Dance hall (Ing. Agripa Popescu hall) – without ventilation Point #2		37	10.4	1008.5	78	0.6
3	Dance hall – with ventilation Measuring point #3		37	11.5	1009	75	1.4
4	Gym – without ventilation Point #4		63	10.5	1009	75	0.5
5	Gym – with ventilation Measuring point #5		63	11.1	1009	75	1.4
6	Access point into the salt mine Point #6		-	21	1009.6	74	1.6

Table 2

The measuring points with coordinates and environment conditions

No.	MEASURING POINT/LOCATION	AVERAGE RADON CONCENTRATION [Bq/m^3]	S(n-1) [Bq/m^3]	S(n-1) [%]	S(med) [Bq/m^3]	S(med) [%]	S(Poisson) [%]	Error [Bq/m^3]
1	Salt lake (swimming pool Point: #1	96.5	17.37	18	2.78	2.88	1.73	4.76
2.	Dance hall – non vent Point #2	94.9	11.36	11.97	1.84	1.94	1.77	4.22
3.	Dance hall – vent Point: #3	50.8	9.62	18.94	4.30	8.47	6.54	4.76
4.	Gym – non vent Point: #4	93.8	8.85	9.44	3.61	3.85	4.48	5.21
5.	Gym – vent Measure point #5	90.2	10.45	11.58	3.69	4.09	3.95	5.16
6.	Access point into the salt mine Point: #6	20.5	2.03	9.90	1.01	4.95	10.85	1.30

Point: #1 - Salt lake **(indoor-non vent)**

Average =(96.5 ± 4.76) Bq/m^3

Table 3

S[n-1]	17.37
S[n-1]%	18.00
S[med]	2.78
S[med]%	2.88
S[Poisson]%	1.73
Err[Bq/m^3]	**4.76**

Point: #2 - Dance hall (**indoor – non vent.)**

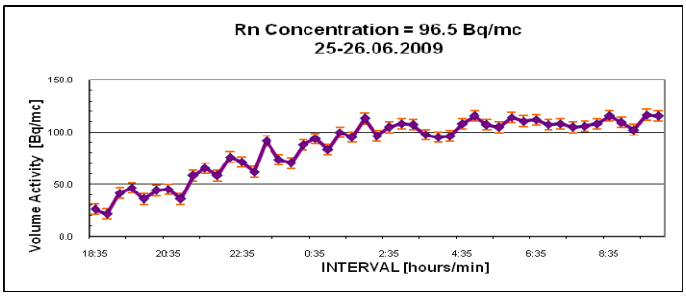

Fig. 1

Average =(94.9 ± 4.22) Bq/m^3

Table 4

S[n-1]	11.36
S[n-1]%	11.97
S[med]	1.84
S[med]%	1.94
S[Poisson]%	1.77
Err[Bq/m^3]	**4.22**

Point: #3 - Dance hall

(indoor - vent.)

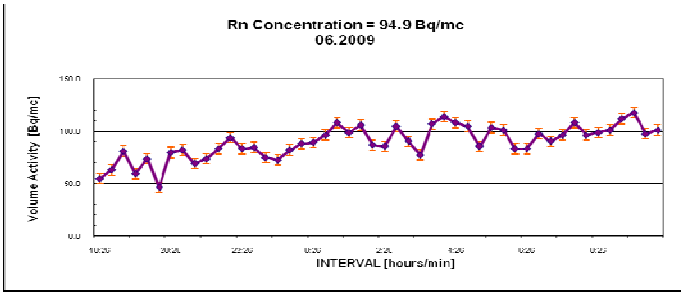

Fig. 2

Average = (50.8 ± 4.76) Bq/m^3

Table 5

S[n-1]	9.62
S[n-1]%	18.94
S[med]	4.30
S[med]%	8.47
S[Poisson]%	6.54
Err [Bq/m^3]	**4.76**

Point: #4 - **Gym**

(indoor - non vent.)

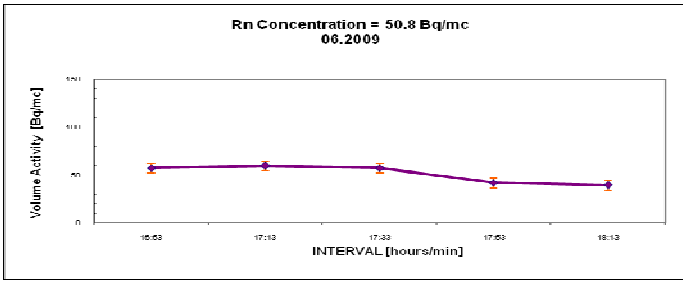

Fig. 3

Average = (93.8 ± 5.21) Bq/m^3

Table 6

S[n-1]	8.85
S[n-1]%	9.44
S[med]	3.61
S[med]%	3.85
S[Poisson]%	4.48
Err [Bq/m^3]	**5.21**

Point: #5 - Dance hall

(indoor - vent.)

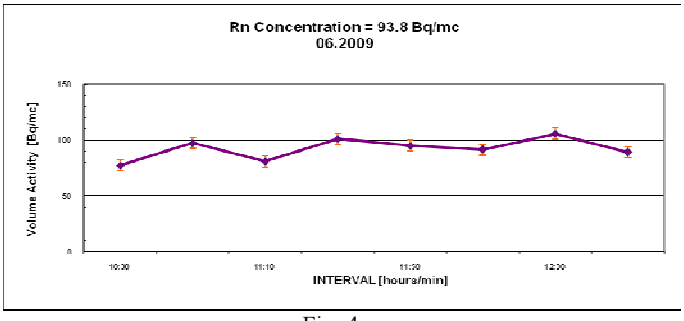

Fig. 4

Average = (90.2 ± 5.16) Bq/m^3

Table 7

S[n-1]	10.45
S[n-1]%	11.58
S[med]	3.69
S[med]%	4.09
S[Poisson]%	3.95
Err%	**5.16**

Point: #6 – **Access point into the salt mine (outdoor)**

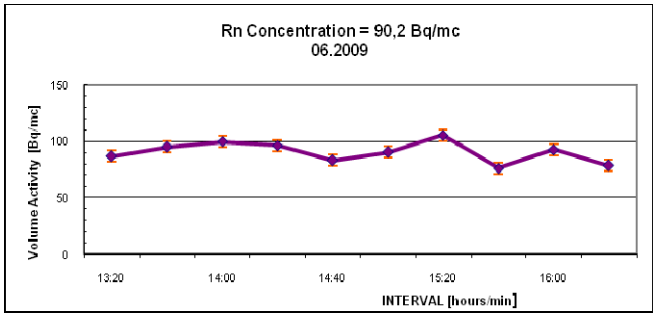

Fig. 5

Average = (20.5 ± 1.30) Bq/m^3

Table 8

S[n-1]	2.03
S[n-1]%	9.90
S[med]	1.01
S[med]%	4.95
S[Poisson]%	10.85
Err%	**1.30**

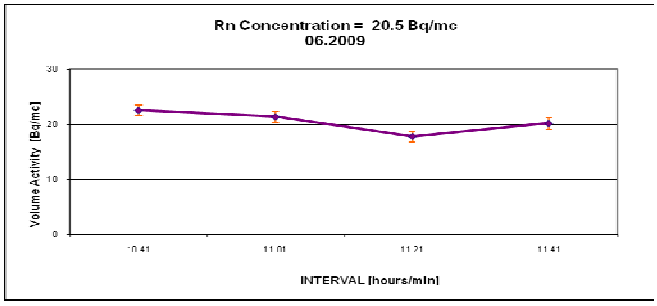

Fig. 6

2.2. GAMMA SPECTROMETRIC MEASUREMENTS ON SALT SAMPLES WITH GEHP DETECTOR

The experimental measurements were made using a *ORTEC* detector with *GeHP* with the following installation parameters: time constant of 6 μs with a relative efficiency of 30 % at 1 332 keV for ^{60}Co and a resolution of 1.85 keV at 1 332 keV (^{60}Co), and 0.85 keV at 122 keV (^{57}Co), work tension + 4 400 V and a spectrometric *DigiDART* chain.

The spectrums were acquired in the energetic domain ranging between 40 and 2 610 keV, on 16 384 channels so as from an energetic point of view a channel corresponds to appreciatively 0.16 keV [3, 4]. The experimental measuring data are presented in Table 9 using the Gamma *Vision-32 (ORTEC)* software of the measuring system on samples of rough salt with a mass of 100 g taken from the Cacica salt mine and measured for 86 400 s (time live) [5].

The principle of the data processing is the following: during the analysis of the obtained ROIs for the same radionuclids, the activities have been deduceted line by line and then an average was calculated.

From this analysis resulted that the most acurate are the 15 peaks of the ^{214}Bi. It can be observed that the value of the activity rises as the energy of the peak rises. This happens when, due to density differences between the sample and the ethalon source, the absorbtion grows when the energy is low, leading to the evaluation of the dependence activity of the analised peak.

The decrease of the calculated activity *peak to peak* between the same energies of the background and of the samples eliminates almost completely this discrepancy. After analyses done on several work files it can be observed on the Figs. from the *SALT_CACICA_with 2 AVG_GRAFERR_with_BARS.XLS* folder (Figs. 9, 10, and 11) that the weighted average shows final errors less significant than the direct average. The weights are the inverse squares of the statistical errors given by the analysis of the ROIs by the program. There errors are in general composed by the statistical error of the net surface of the respective peak. All the errors taken into account are relative errors. As it can be observed in Figs. 9, 10 and 11 the weighted averages give smaller final errors and thus only they were taken into consideration, containing the same data but having a different graphical method of solving. From the graphics presented in the Excel file *SALT_CACICA_GRAFERR_with BARS.XLS*, it can be seen which are the radionuclids that appear with the highest probability, while being present in the salt. These are those elements for which the value of the average decrease is positive with an error smaller than the positive value. These are ^{226}Ra, ^{40}K and ^{177}Re K-xray. The rhenium isn't a natural radioactive radionuclide. Its presence in the salt sample is thought to be due to the excitation by x-ray fluoresce of lead's characteristic radiation which is present in the natural radiation background.

Table 9 shows the *ROI REPORT* given by the Gamma Vision software of the spectrometric system on the measuring data of the salt samples. The background spectrum has been deducted and the data from Tables 10 and 11 was processed.

The principal radionuclids are presented with their spectral lines, the differences between the sample's activity and the activity of the background Bq(p) – Bq(f) and the calculation of the direct average and the weighted average etc. [6,7].

For calculating the averages we used the following formulas:

For the direct average:

$$\overline{x} = \frac{x_1 + x_2 + \ldots x_n}{N} = \frac{1}{N}\sum_{i=1}^{N} x_i. \quad s = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(x_i - \overline{x}\right)^2}, \tag{1}$$

And for the weighted average:

$$\overline{x} = \frac{\sum_{i=1}^{N} p_i x_i}{\sum_{i=1}^{N} p_i}, \quad s = \sqrt{\frac{N'\sum_{i=1}^{N} p_i \left(x_i - \overline{x}\right)^2}{\left(N'-1\right)\sum_{i=1}^{N} p_i}}, \tag{2}$$

Table 11

The values of points represented in the following figures comparative for the two averages with the error bars

X	ERROR	Y
Ac-228_(-)>MED	0.500	0.608
_(-)>MEP	0.495	0.344
Bi-212_(-)>MED	0.350	-0.230
_(-)>MEP	0.337	-0.135
Bi-214_(-)>MED	0.869	-0.493
_(-)>MEP	0.375	-0.386
Pb-212_(-)>MED	0.137	-0.152
_(-)>MEP	0.117	0.031
Pb-214_(-)>MED	0.195	-0.933
_(-)>MEP	0.041	-0.620
Re-177Kxray_(-)>MED	0.566	1.494
_(-)>MEP	0.374	2.129
Th-234_(-)>MED	4.820	-3.990
_(-)>MEP	3.737	-0.945
Tl-208_(-)>MED	0.040	-0.018
_(-)>MEP	0.028	0.103

(-)>MED= first subtraction and then direct average
(-)>MEP= first subtraction and then a weighted average
MED = direct average
MEP = weighted average

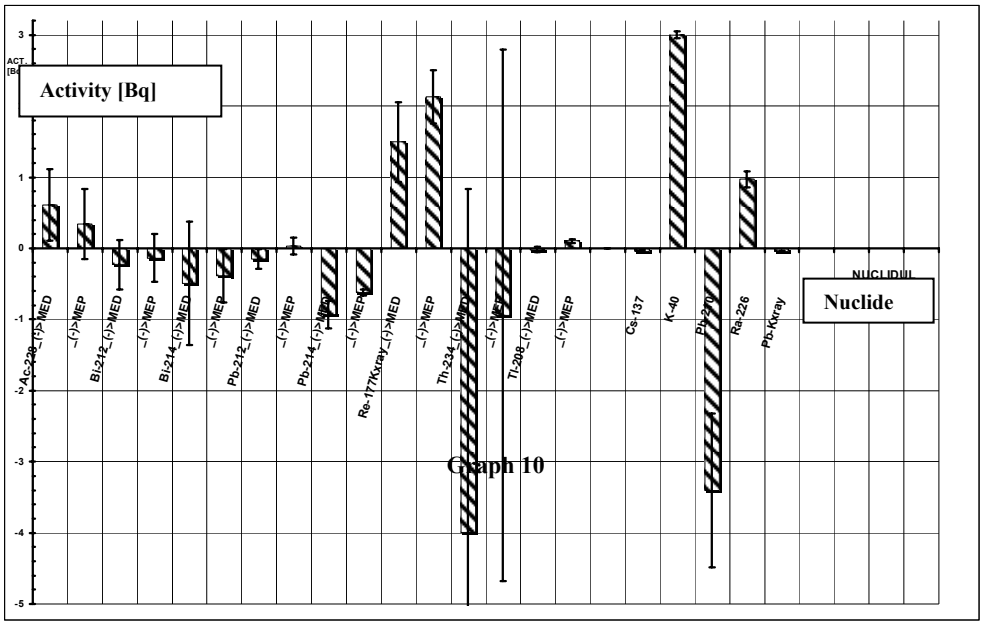

Fig. 9

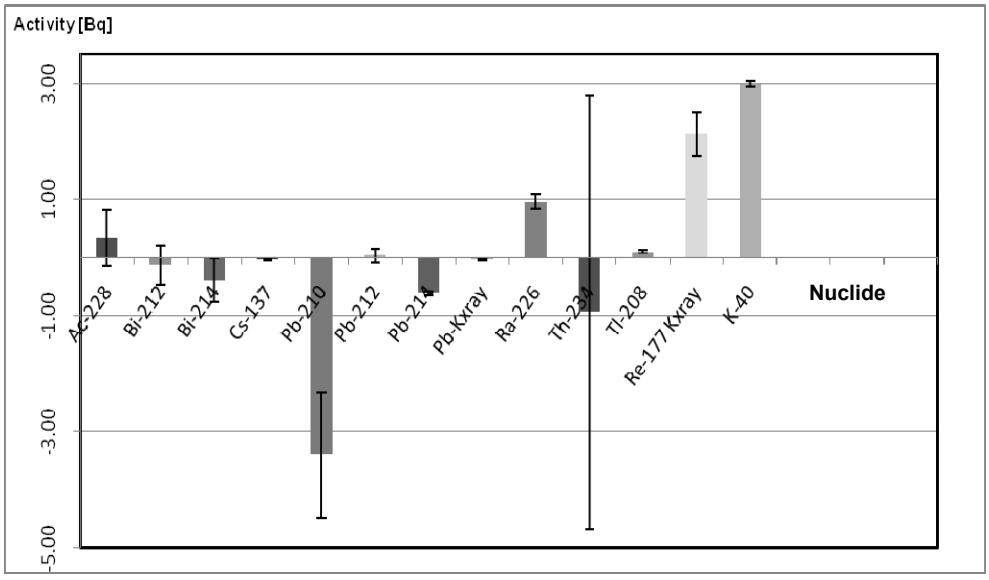

Fig. 10

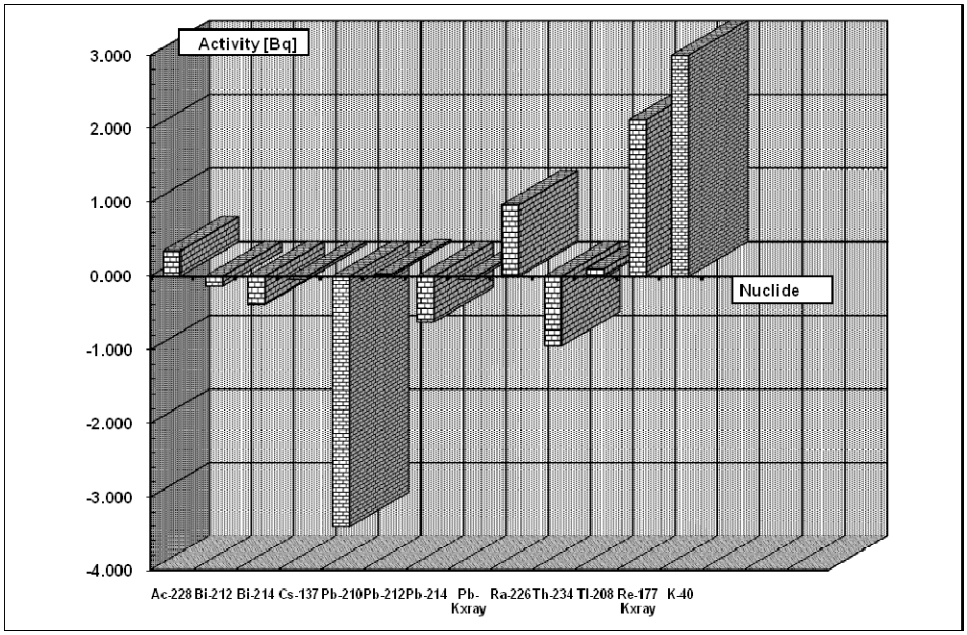

Fig. 11

In the spectrum of samples of salt, one can notice small traces of: ^{228}Ac (the lines of 911,07, 964.6, 968.9, 1587.9, 1630.4), ^{212}Bi (the line of 1620.56), ^{214}Bi (the lines of 609.32, 1120.28, 1155,19, 1238.11, etc. as in table No. 10), ^{137}Cs (the line

of 661.62), ^{40}K (1460.75), ^{210}Pb (46.52), ^{214}Pb (241.92, 295.22, 351.99), ^{177}Rn, ^{226}Ra (185.99), ^{234}Th (63.2, 92.38), ^{208}Tl (510.72, 583.14, 1592.47, etc), etc. all in the natural background radiation (according to Table 10).

2.3. GLOBAL ALPHA-BETA MEASUREMENTS WITH THE MPC 2000 DP SYSTEM

Unprocessed salt samples, with a mass of 1g and 2g, taken from the salt mines have been submitted to alpha-beta global measurement/testing with the MPC 2000 system, in <*ALPHA+BETA SUS*> measurement geometry, and experimental measurement data, recorded and automatically generated by the system, are presented in Table 12, after which are calculated: $S(n-1)$ – experimental standard deviation, $S(n-1)$, [%] – relative experimental standard deviation, S(med) – experimental standard deviation of the mean, S(med), [%] – relative experimental standard deviation; S(Poisson) – Poisson relative error, global absolute error, etc. The effectiveness of alpha radiation detection (AEFF) is 36.23%, the effectiveness of beta radiation detection (BEFF) is 45.72% and minimum detectable activities:

A_AMD for alpha = 0.0474 Bq;

B_AMD for beta = 0.394 Bq.

Average activity for alpha radiation = A_ACT= 0.007963 Bq;

Average activity for beta radiation = B_ACT=-0.053483 Bq.

For salt samples taken, the mean values of specific global beta activity were below the warning limit of 50 Bq/m^3 established by legislation (*MAPM Ministerial Order* no. 338/2002) [8, 9].

Table 12

Experimental data generated by the *MPC 2000* system calculated and processed

ALPHA ACTIVITY	STANDARD DEVIATION ALPHA ACTIVITY	MINIMUM DETECTABLE ALPHA ACTIVITY	BETA ACTIVITY	STANDARD DEVIATION BETA ACTIVITY	MINIMUM DETECTABLE BETA ACTIVITY
[Bq]	[Bq]	[Bq]	[Bq]	[Bq]	[Bq]
-0.00506	0.00048	0.0474	0.10200	0.117	0.394
0.00414	0.00921	0.0474	-0.03860	0.113	0.394
-0.00506	0.00048	0.0474	0.05850	0.116	0.394
0.02250	0.01590	0.0474	-0.02120	0.113	0.394
0.03170	0.01840	0.0474	-0.23500	0.106	0.394
0.00414	0.00921	0.0474	-0.02400	0.113	0.394
0.00414	0.00921	0.0474	0.07080	0.116	0.394

Table 12 (continued)

0.02250	0.01590	0.0474	-0.07230	0.112	0.394
0.00414	0.00921	0.0474	-0.27200	0.105	0.394
0.00414	0.00921	0.0474	0.09270	0.117	0.394
0.00414	0.00921	0.0474	-0.23500	0.106	0.394
0.008311	**Bq**		**- 0.052191**	**Bq**	
142.96022	**S (n-1)[%]**		**263.86213**	**S (n-1)[%]**	
41.269061	**S (med)[%]**		**76.170435**	**S (med)[%]**	
± 0.003429	**Bq**		**± 0.039754**	**Bq**	

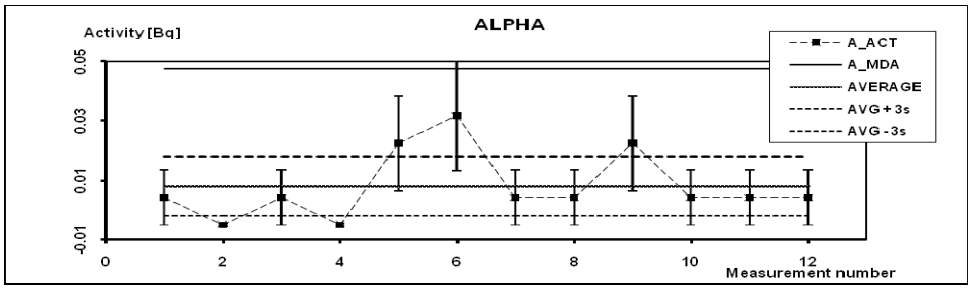

Fig. 12

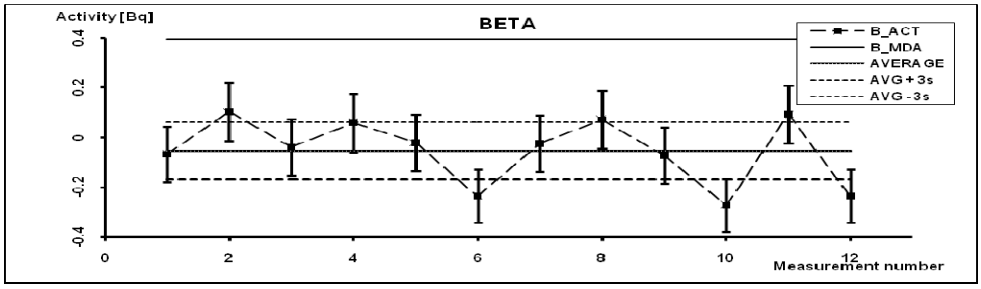

Fig. 13

Figures 12 and 13 shows that all values and averages with 3 times the standard deviation are below the minimum detectable, both for the alpha radiation, and the beta radiation.

2.4. MEASURING THE NATURAL BACKGROUND RADIATION

Measuring the range of natural background gamma radiation is one of the most laborious measurements because it requires very sensitive equipment, which generally involves large volume detectors. Such equipment is very rare and usually

non-transportable, which is why we used a typical flow meter for measurements in the salt mine. For such a flow meter, international regulations are applied, that establish the parameters that must be born, and the maximum permissible uncertainties.

We have used this dose equivalent flow meter, under the lower limit of the measurement range, meaning that range for which the value displayed enters the allowed uncertainty in maximum 8 seconds. For this reason, we had to use one of the device's features, in order to increase the statistic accuracy of the number of impulses at very low counting rates, such as those of a salt mine, that is below ground level, by integrating the long-term detected impulses. We selected the operating mode *Counter-timer* to perform stationary measurements with a *default precision (accuracy) for the average value*. In this way, either the *mediation period*, or – by presenting the pulses – the *statistical precision (accuracy)* of the measured value can be predetermined. If very high accuracy is desired, the fund may also be determined in the CPS (raw data) operating mode with counter-timer (directly before the respective measurement). The measuring of the radiation fund in the Cacica salt mine was achieved with the Berthold Umo LB 123 portable system, which is also a dosimeter/debitmeter and contaminometer. Measurements in the salt mine were obtained with the integrated impulse debitmeter using the gamma probe – *Counter-timer* for integration times of 3 600 seconds.

It follows from the measurement data that background radiation is: in the salt mine: 0.016 μSv/h (0.143 CPS), and on the surface: 0.127 μSv/h (0.658 CPS), with a statistic that ensures an error of 5.2%.

So, the background radiation in the salt mine is about 8 times smaller than on the surface.

2.5. MEASUREMENT OF TRITIUM IN THE CACICA SALT MINE

Collection of tritium present in the form of tritium water in the atmospheric air in the Cacica salt mine was accomplished with the *TROTEC TTK 100S* dehumidifier placed at a time at different points in the salt mine. Both the collected condensate and water samples from the groundwater were processed at the Radiochemistry Laboratory for Environmental Samples in the *Department of Life and Environmental Physics of IFIN-HH* by distillation at atmospheric pressure and measuring with the Tricarb 1600TR liquid scintillation spectrometer, according to current procedures [10-19]. The experimental results obtained are summarized and explained in Table 13.

The value associated to the fund was established by routine tests for "reference water". In this respect, an averaging was achieved for measurements of background samples for the period of analysis, estimated for 3 vials. The "reference water" used is *Dorna flat water* (depth spring). The average used in the above calculations was 5.33 cpm (standard deviation 2σ – 8.66%, detection efficiency – 0.24057).

Table 13

The level of tritium present in the form of tritium water in the water and atmospheric air in the Cacica salt mine

No.	Location of sampling	Date of sampling	The volume of water taken, mL ≅ The amount of water, [g]	$A \pm \sigma_A$ (Bq/dm^3)	Observations
1.	Dance hall	24.06.09-25.06.09	250	3.86 ± 0.5	11 h 30 min of sampling; t = 11^{o}C AMD = 5.5 Bq/dm^3 A < AMD
2.	Gym	25.06.09	350	4.74 ± 0.6	16 h of sampling; 11.6^{o}C AMD = 5.5 Bq/dm^3 A ≈ AMD
3.	Swimming pool water (lake)	26.06.09	400	0.80 ± 0.1	AMD = 5Bq/dm^3 A < AMD
4.	Pond water	27.06.09	500	Sub fond	AMD = 5Bq/dm^3 A < AMD

3. CONCLUSIONS

• Measurements made at the Cacica salt mine were radioactive radiation measurements for alpha, beta and gamma radiation, atmospheric radon and tritium;

• In situ measurements were natural radiation background measurements and measurements of atmospheric concentrations of radon inside the mine, but also at the surface of the mine/control measurements;

• Laboratory measurements were performed on samples of raw salt and water, taken from the salt mine, and all analyzed/measured samples of salt, air, water are safe in terms of radioactivity and the values obtained show that it is a clean environment, with limited contact with air at the surface;

• The values obtained for tritium in atmospheric air are higher than those for underground lakes; this is explained by the fact that there is exchange of air with the surface; the value in the gym is higher than that obtained for the dance hall;

• When ventilation is functional, concentrations of Rn and tritium decrease quite a lot, compared to the nonaeration situation.

• The values obtained for the lake waters show that they are very clean and come from deep water, and not from infiltration or surface groundwater; All values obtained are around the AMD value; the resumption of testing with a device to measure very low levels (*Quantulus 1220*) is recommended and possibly enrichment by electrolysis;

• The Cacica salt mine is a very clean environment, unadulterated and free of other human industrial activities, which prevents cosmogonic production of radioactive impurities and is required for regular study of the radioactive background, as well as repeating the dose and low-level dose flow measurements, evaluating fluctuations in the integral background counting rate, environmental studies using other systems with very low detection limits for long measurement times.

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