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MONITORING OF THE RADIOACTIVITY CONCENTRATION OF AIR IN THE AREA OF THE IFIN-HH, ROMANIA*

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Abstract. The paper presents the concept and the results obtained during an annual survey of the radioactivity concentrations in our Institute, a nuclear centre, involving research, production and use of radioisotopes, a nuclear reactor under decommissioning and a radioactive wastes treatment unit. The Department of Life and Environment Physics, in charge with the survey, established procedures for sampling , measurement and reporting of the results. Generaly, for the air survey the sampling is based on the use of air filters. The measurements are carried out at established time intervals chosen such as to cover the measurement of the short lived natural radon daughters, as well as the monitoring of long lived natural and artificial radioisotopes. The measurements are performed by the following methods: (i) gross alpha and beta activity measurements 3 minutes after collection, after 20 h, and after 5 days, by the use of a high sensitivity gross alpha-beta activity system; (ii) gross gamma activity after 5 days from prelevement, by the use of a gamma measurement system. The results obtained are in good agreement for the various methods. At the same time, the conclusion is that the main part of the measured radioactivity is due to the radon short life daughters: lead and bismuth, such as the measurements performed at various intervals showed. The content of the artificial radionuclides is situated within the same interval as in other Romanian areas where no nuclear activitie are deployed.

Key words: air radioactivity concentration, survey procedures, radon daughters.

1. INTRODUCTION

The radioactive content of air is continuously monitored on the placement of the IFIN-HH area, due to the existence of some units, such as the Radioisotope Center and the Department for Radioactive Wastes Treatment, where open radioactive sources are manipulated. Although all the measures are taken, in order to avoid any elimination of radioactive gaseous effluents over the established limits in atmosphere, a permanent survey is necessary, in order to adequately limit the radiological hazard in the shortest time.

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This survey is carried out by two parallel methods: (i) direct measurement of radioactive concentration of air by the method of retaining the aerosols in a filter paper; (ii) survey of the ambient dose equivalent rate all the year long. The results of the two methods are correlated, as the dose rate value is influenced by the radioactive concentration of air. Some discrepancies could occur near the buildings operating with high activity sealed sources, not adequately shielded, the movement of such sources within the IFIN-HH area, or detection of some orphan sources. The methods and equipment for the measurement of radioactive concentration of air are described in detail in the paper. The measurement results for the year 2006 survey, with their discussion are also presented.

2. RADIOACTIVE CONCENTRATION OF AIR (ACTIVITY DENSITY) MEASUREMENT

2.1. COLLECTION OF SAMPLES

The method is based on the collection of aerosols, which are supposed to contain radioactive fractions attached to them. The method is standardized and relies on the use of an aspiration pump, type EγJDL28BE, with an well determined air flow rate, 66 to 70 L/min. The aspiration is done within pre established periods of time, usually 5 hours, the total volume of air varying between 19,800 and 21,000 L. The aspiration is done through an air filter, type NO 521147, with a porosity 1μm, which assures a retention yield of 96–99%. The aspiration point is placed at a height of 16 m from the soil. The aspirations of air are performed every two weeks, all the year long. A special care is paid to the filter examination for assurance of the yield prescribed, as this parameter influences directly the calculations: the filter must not present non uniformities, must not be folded before use, and must be manipulated only by using pincers. The atmospheric parameters are: temperature range: -20^0C…42^0C; pressure: 710 …765 mmHg.

2.2. MEASUREMENT OF ACTIVITY

As almost all the air contaminants are alpha, beta-gamma emitters, the measurements of air filters are performed by the measurement of these radiations. It is well known that the aerosols can contain two types of contaminants: (i) a contamination with ^{222}Rn and ^{220}Rn daughters, heavy metals, ^{212,214}Pb, ^{212,214}Bi, ^{214,216,218}Po; (ii) artificial radionuclides, such as long half life fission products: ^{90}Sr, ^{137}Cs , short half life, immediately after an accident: ^{99}Mo, ^{103}Ru ,^{131}I, or effluents discharged during the operation of nuclear installations.

The method was adapted such as to make distinction between the natural and artificial components of contamination. From UNSCEAR published data, the concentration of Radon isotopes, at the world scale, over the terrestrial area, is: ^{222}Rn – 10 Bq·m^{-3}; ^{220}Rn – about 1 Bq·m^{-3} [1]. The reported mean values in Romania are similar [2]. ^{222}Rn decay ($T_{1/2}$= 3.8235 d) results in short half life alpha and beta-gamma emitters: ^{218}Po ($T_{1/2}$ = 3.05 min), ^{214}Pb ($T_{1/2}$ = 19.9 min), ^{214}Bi ($T_{1/2}$ = 26.8 min); ^{220}Rn decay ($T_{1/2}$ = 55.6 s) results in: ^{216}Po ($T_{1/2}$ = 0.15 s), ^{212}Pb ($T_{1/2}$ =10.6 h), ^{212}Bi ($T_{1/2}$ =60.6 min). Two types of measurements are performed.

2.2.1. Measurement of alpha, beta radiations

The measurement of alpha and beta radiations is made according to the Romanian Standard STAS 12457-86 [3] by the use of the Alpha-Beta global Analyzer, type PROTEAN, model MPC-9300, specially designed for low level activity measurements, having the background counting rates: 0.04 min^{-1} for alpha, and 0.5 min^{-1} for beta radiations. The method consists in the followings. After the expiry of the aspiration time, 5 hours, the filter is removed from the aspiration pump and measured 3 minutes after, for a 10 min registration time. Parallel, alpha and beta radiations are registered, and corrections for background contribution are operated; the following measurement is done after a 20 hours time, resulting the corrected counts. It is considered that at this time, the contribution of ^{222}Rn daughters is zero, and is due only to ^{220}Rn ones. The last measurement is done after 5 days, when all ^{220}Rn daughters decayed, only artificial radionuclides having contributions; counting rates, $R_{3\alpha}$, respectively $R_{3\beta}$ are registered. Due to the significant diminution of the counting rate, 50 minutes measurement times are used.

The formulae for the calculation of the individual contributions of ^{222}Rn, ^{220}Rn daughters were taken from the paper [4], which refers to alpha radiations measurements.

$$A_{\mathrm{Pb212}} = \lambda_1 \left[\varepsilon q G \left(T_c, T_3, T_4 \right) \right]^{-1} C_2 ,$$
$$A_{\mathrm{Rdn}} = \lambda_3 \left\{ \varepsilon q [F_1 (T_c, T_1, T_2) + F_2 (T_c, T_1, T_2] \right\}^{-1} (C_1 - C_2'). \quad (1)$$

In relations (1), A_{Pb212} refers to ^{220}Rn (Thoron) daughters, consisting mainly from ^{212}Pb, and A_{Rdn} refers to ^{222}Rn daughters. λ_i refers to the decay constants of the daughters: ^{212}Pb, ^{212}Bi, ^{218}Pb, ^{214}Pb, ^{214}Bi , respectively, expressed in s^{-1}. G, F_1, F_2 are radioactive decay expressions, written by the cited authors. T_c, T_1, T_2, T_3, T_4 expressed in seconds, represent: collection time, times from collection time to the measurement, and measurement times. $\boldsymbol{\varepsilon}$ are detection efficiencies, in our case: ε_α = 0.48 s^{-1}Bq^{-1}, ε_β = 0.31 s^{-1}Bq^{-1}. C_1, C_2, C_2' represent the numbers of counts

registered during the measurement times, 10 min (after 3 min from collection time) and 50 min (after 20 h). q represents the pump air flow rate, $m^3\ s^{-1}$.

The relation for concentration of artificial radionuclides, Cs-137, I-131, Sb-125, Cs-137, Sr-90 according to [3], is:

$$A_{artificial} = \frac{R_3}{V \eta f_{et}} \cdot . \tag{2}$$

A are radioactive concentrations (activity densities) expressed in units*:* Bq m^{-3}. R_3 are expressed in min^{-1}; V is air volume, expressed in m^3; η is the retention yield of the filter, 96%; $\mathbf{f}_{et} = \varepsilon_\beta$ is the calibration factor, expressed in $min^{-1}Bq^{-1}$.

The evaluation of uncertainty is calculated by the quadratic summation of all individual components: type A (statistic uncertainty), u_A, and type B components due to uncertainties of the parameters from relations (1) and (2). In the case of very low concentrations, one evaluates first the minimum significant R_3 counting rate, and the minimum detectable activity is calculated according to:

$$A_{md} = \frac{R_{md}}{V \eta f_{et}} \left[\mathrm{Bq} \times \mathrm{m}^{-3} \right]. \tag{3}$$

R_{md} is determined from the fluctuations in the background counting rate. The reported value is then:

$$A < A_{\mathrm{md}}. \tag{4}$$

2.2.2. Measurement of gamma-rays

A gross gamma-ray measurement is made after 5 days from the sample collection time, in order to detect possible artificial gamma emitters. The gamma measurement system is provided with a well type NaI(Tl) crystal, adequately shielded, where the filter foils are introduced after performing gross alpha and gross beta measurements, in order to get a high detection efficiency.

3. RESULTS AND DISCUSSION

3.1. MEASUREMENT DATA

The summation of all gross alpha, beta and gamma measurements performed during the year 2006, calculated according to relations (1) for natural radionuclides, columns1 – 4, and according to relation (2) for artificial radionuclides, columns 5 and 6, is presented in the next table Table 1.

Table 1

The results for measurements of radioactivity in air

Month	Gross Alpha measurement after 3 min, A_{Rdn} [Bq m^{-3}]	Gross Alpha, measurement after 20 h, $A_{Pb\text{-}212}$ [Bq m^{-3}]	Gross Beta, measure ment after 3min, [Bq·m^{-3}]	Gross Beta, measurement after 20 h, Bq·m^{-3}	Gross Beta, measurement after 5 days [Bq·m^{-3}]	Gross Gamma, measurement after 5 days [Bq·m^{-3}]
0	1	2	3	4	5	6
01	2.07±0.21	0.074±0.020	2.14±0.09	0.120±0.05	0.004±0.0015	0.0052±0.002
01	2.14±0.21	0.079±0.020	1.44±0.07	0.058±0.005	0.006±0.0013	0.0045±0.0014
02	4.37±0.44	0.075±0.020	3.03±0.13	0.056±0.003	0.0047±0.0012	0.0051±0.0022
02	2.29±0.23	0.180±0.060	1.96±0.09	0.020±0.009	0.0077±0.0032	0.0074±0.0025
03	2.03±0.20	0.064±0.020	1.74±0.08	0.053±0.006	0.006±0.0024	0.0073±0.0032
03	2.19±0.22	0.089±0.030	2.19±0.10	0.098±0.008	0.006±0.0025	0.0078±0.0032
04	2.12±0.21	0.120±0.040	2.14±0.10	0.150±0.009	0.006±0.003	0.0074±0.0032
04	1.98±0.20	0.124±0.040	1.97±0.02	0.020±0.003	0.006±0.0022	0.0075±0.0032
05	2.08±0.21	0.060±0.020	1.65±0.08	0.060±-.006	0.008±0.0034	0.0069±0.0034
05	2.07±0.21	0.104±0.030	2.51±0.11	0.170±0.009	0.006±0.003	0.0075±0.0042
06	1.29±0.13	0.097±0.030	1.02±0.05	0.080±0.007	0.006±0.0025	0.0065±0.0033
06	2.38±0.24	0.120±0.036	2.78±0.12	0.020±0.009	0.010±0.004	0.0077±0.0043
07	1.83±0.18	0.091±0.030	1.62±0.08	0.090±0.007	0.006±0.0025	0.0068±0.0032
07	2.95±0.30	0.200±0.060	1.57±0.08	0.30±0.10	0.008±0.004	0.0072±0.0032
08	1.84±0.18	0.071±0.020	2.28±0.10	0.057±0.006	0.007±0.003	0.0074±0.0045
08	3.79±0.38	0.140±0.050	3.14±0.14	0.140±0.07	0.005±0.003	0.0069±0.0035
09	3.12±0.31	0.220±0.070	2.79±0.12	0.250±0.10	0.006±0.003	0.0067±0.0033
09	2.59±0.26	0.244±0.070	3.11±0.18	0.280±0.08	0.008±0.003	0.0069±0.0012
10	3.44±0.34	0.169±0.050	3.03±0.14	0.170±0.009	0.007±0.0008	0.0063±0.0034
10	5.95±0.60	0.41±0.12	5.04±0.18	0.370±0.17	0.010±0.004	0.0094±0.0036
11	2.82±0.28	0.246±0.070	3.51±0.15	0.310±0.16	0.008±0.003	0.0069±0.0036
11	5.18±0.52	0.168±0.050	5.34±0.22	0.350±0.16	0.0076±0.004	0.0067±0.0044
12	12.03±1.2	0.455±0.13	11.45±0.45	0.590±0.25	0.0077±0.003	0.0065±0.0032
Mean values	3.15 ±2.24	0.16± 0.10	2.93±2.13	0.17± 0.09	0.0068 ± 0.0015	0.0069 ± 0.0010

3.2. DISCUSSION OF RESULTS PRESENTED IN TABLE 1

The ^{222}Rn concentrations in air such as presented in column 1, gross alpha measurements, have values from 1.3 to 12.0 Bq·m^{-3}, with a mean value of (3.15± 2.24) Bq·m^{-3}. The parallel beta measurements, column 3, are in agreement with alpha determinations. The ^{220}Rn daughter concentration in air, presented in column 2 for alpha measurements varies from 0.07 to 0.46 Bq·m^{-3} and a mean of (0.16±0.10) Bq·m^{-3}. The parallel gross beta measurements, presented in column 4 agree with values from column 2, individually and as mean values. The well known annual variation trend is observed, and the mean ratio of the two series of daughters [A_{Rdn}/A_{Pb212}] = 0.05. All these values are roughly in agreement with other reported data [1, 2]; maybe they are a little lower, due to the height of the collection point, 16 m.

Regarding the values measured after 5 days, considered as contributions from artificial radionuclides, one may conclude. The measurements by gross alpha method showed in almost all cases that the activities were less than minimum detectable activity. It is considered that none alpha emitting radionuclide is detected in air over the IFIN-HH area. Regarding the gross beta measurements, the values presented in column 5, lie between 0.0040 and 0.0096 $Bq \cdot m^{-3}$, with a mean value (0.0069±0.0015) $Bq \cdot m^{-3}$. The comparative measurements, performed by the gross gamma method, column 6, are in good agreement with those presented in column 5. This agreement may be interpreted as none pure beta emitters, such as ^{90}Sr, is present in air. These values, situated over the detection limit, are supposed to be due to the artificial beta-gamma long half life radionuclides. If one compares these values with the fixed limit for gaseous effluents, as high as 3.7 $Bq \cdot m^{-3}$ one may conclude that these concentrations are much lower, situated at 0.2% from the legal limit.

CONCLUSIONS

A complete procedure for collection, measurement and processing of data, all the year long, was implemented in the IFIN-HH, Bucharest, Romania.

The use of two parallel methods allowed to verify the validity of methods and to report confident results to the decisional bodies.

The main part of radioactive content of air is due, as expected, to ^{222}Rn and ^{220}Rn daughters; our results are in line with literature data.

None artificial alpha emitter was found, and the content of beta-gamma emitting radionuclides is situated at a level of 0.2% from the legal limit for gaseous effluents.

REFERENCES

1. B.G. Bennet, *Assessments by UNSCEAR of Radiation Sources and Effects*, IRPA 9 Congress. Proc., Vol. 1, p. 35, Vienna, Austria, 1996.
2. Societatea Română de Radioprotecție, *Radioactivitatea naturală în România* (in Romanian), Bucureşti 1994.
3. Romanian Standard, STAS No. 12457–86, *Determinarea densității de activitate a aerosolilor* (in Romanian), 1986.
4. T. Kataoka, E. Yunoki, K. Michihiro, H. Sugiyama, M. Shimizu, T. Mori, O. Tsukamoto, K. Sahashi, *Simultaneous and automatic determination of ^{222}Rn daughters and ^{212}Pb concentration in the atmosphere using two α scintillation counters*, IRPA 9 Congress Proc., Vol. 2, p. 14, Vienna, Austria,1996.