Study Of Indoor Radon, Thoron, And Annual Effective Doses Using Twin Chamber Dosimeter Cups In The Dwellings Of Mathura (U.P.), India.

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ABSTRACT:- Mathura is a city in the north Indian prefecture of Uttar Pradesh. Mathura oil processing plant sited in the city is one of the utmost petroleum treatment facilities in Asia which was built up in 1978. The Mathura petroleum processing plant discharges harmful gases and arranges squander into the waterways making the region filthy and undesirable. In the present study attention has been given to the estimation of radon and its descendants to know the effect of radioactivity in nearby homes due to refinery waste. Radiological significance of radon and its descendants have been recognized quite a while back, thoron had frequently been disregarded because of its short half-life. Numerous investigations demonstrate that Thoron and its progeny is likewise a noteworthy contributor to the Residential buildings in some Asian and American nations. The present investigation was completed using Solid State Nuclear Track Detectors (SSNTD's) based twin chamber dosimeters. Radon concentration are found to vary from 35.1 ± 4.5 Bq m^{-3} to 84.8 ± 7.0.Bq m^{-3}, whereas thoron concentrations vary from 3.5±0.3 Bq m^{-3} to 24.3±2.2 Bq m^{-3}. The annual effective dose from radon and thoron was found to vary from 0.10mSv y^{-1} to 0.24mSv y^{-1} which was less then global average value of 1mSv y^{-1}

Keywords: -SSNTD, Radon, Thoron, annual effective dose, Twin chamber dosimeter, spark counter

1 INTRODUCTION:

Naturally occurring radionuclide are substantial indoors the contiguous. Many geological formations incorporate uranium, radium, radon and different radioactive elements collectively with oil and gas. Whilst oil and gas are produced, traces of these radioactive rudiments produced [1], [2]. Over the years radioactive elements are deposited in the scale and sludge, hence contaminating gadgets, vessels, and other centers within the industry and surrounding. In fluctuating degree of seriousness, NORM sullying may exist at each oil and gas creation site and related offices including channel dealing with yards, metal recovery territories, flammable gas and NGL pipelines, gas plants, and NGL processing plant and terminals [3]. There are three forms of NORM contamination inside the oil and fuel industry which show elevated radioactive contamination. Radioactive scale contains uranium, thorium, radium, and related decay products from the generation of oil and related saline solutions sullied with NORM. The radioactivity in the scale begins primarily from radium, which co-hastens with barium and strontium sulfate. Different isotopes in the uranium-238 and thorium-23 decay series may likewise be available. Scale may contain radium-226 in focuses up to 100,000 picocuries (pCi)/gram [1].. Radioactive stream limiting scale might be found in down hole tubing, just as in over-the-ground preparing and transport hardware.

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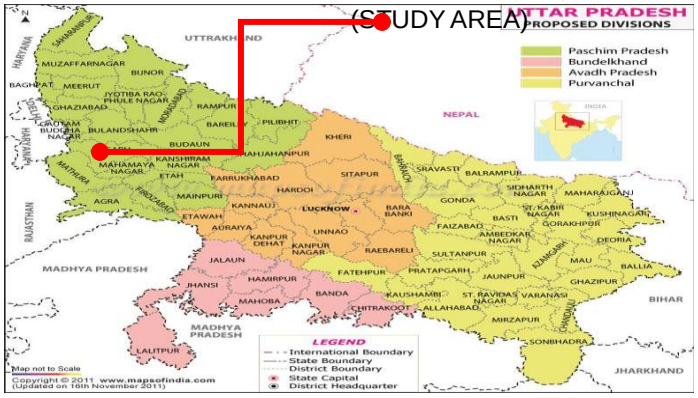
Sullied with NORM might pipe, slime pits, channels, brackish water transfer/infusion wells, and related hardware Likewise, soils defiled from well tubing walkovers-both at the well site and at remote pipe cleaning yards-might be debased with NORM. Radioactive films, coatings, or plating can shape from flammable gas creation sites. Frequently undetectable to the unaided eye, these films contain radon and its decay products regularly with no radon forerunners related with them. Because of radon defilement in petroleum gas, these radioactive films can be found at gas wellheads, in transport funneling, headers, treated units, and siphons, and inside gaseous petrol handling plants or other light hydrocarbon facilities. Radioactive sludge in pipelines, handling plants, NGL stockpiling tanks and conveyance facilities, pigging tasks, and gas lines and other channel gatherings can be tainted with radon in the gaseous petrol. Sludges might be contaminated with a few thousand pCi/gram, Baird et al. 1990 of the enduring radon decay products, i.e., lead-210, bismuth-210, and polonium-210. These overwhelming metal decay products may connect to dust particles and pressurized canned products to turn out to be a piece of the slop. In gas lines, channel congregations expel the radon decay products from the gas together with other particulate issue. Radioactive scale in the oil and gas industry has been accounted for as a prominent special case of a 1975 report by Gesell. NORM defilement of gas facilities by radon and its decay products has not been broadly announced. Radon has been recognized to be a contaminant of natural fuel for nearly one hundred years ago. It was only in 1971 that radon become discovered to concentrate inside the lighter natural gas liquids during processing and will present an extreme health threat to industry personnel, especially to protection personnel. Uranium and radium are available in many soils and shakes in broadly varied concentration, and consequently radon gas is generally dispersed in the earth's crust. Since it is a noble gas and is highly non-reactive. Once framed, radon is allowed to move as a gas, or can dissolve in water, without being trapped or evacuated by chemical reactions. Relocating all through rocks and soil, radon is delivered with gaseous petrol at the wellhead. Radon-222 decays to an atom of lead-210 and consequently to bismuth-210 and polonium-210

preceding rotting to stable lead-206. These extensive radon decay products present a developing issue to the industry, particularly to faculty who might be presented to debased surfaces, mucks, and other waste materials. Polluted offices and waste material issues must be perceived and tended to. The radiations that the materials produce are effectively consumed by the walls of the equipment. The diffusion length of radon, i.e., the average distance an atom can move through dry soil before decaying, is about 1.6 m, while it is only 2 cm for thoron because of relatively short half-life, i.e., 55.6 s. This property of radon makes it a greater health hazardous gas [4]. These radioactive materials are not a wellbeing danger except if they are ingested or breathed in into the body, Whenever breathed in, the residue and pressurized canned products containing NORM can attached to the lung surfaces where they emit alpha particles into the tissue of the lung lining. Investigations of uranium diggers demonstrate that all-inclusive exposure to these radon products poses an expanded danger in the form of lung cancer [5], [6]. Soils have various centralizations of radionuclide relying upon their development from the parent rocks, land area and by geochemical processes [4]. As a latent gas, radon uninhibitedly diffuses through the dirt and achieves the environment where it could move into buildings of abodes to create a wellbeing danger. Inward breath of radon (222Rn) and its fleeting daughter products, particularly 218Po and 214Po are related with the danger of lung disease, if radium content was adequately high and 222Rn was utilized to assess the radioactive peril because of soil and sediment. So it was found to be essential to quantify the dose received by refinery personnel and radon thoron and their progeny deliberation in the contiguous vicinity. In the present work an effort has been made to make indoor radon/Thoron measurements in the some dwellings of Mathura city surrounded Mathura Refinery by using Plastic Twin Chamber Dosimeter cups fitted with SSNTD,s.

2 GEGRAPHY OF THE STUDY AREA

Mathura and Brij Bhumi is the alternative name of Birth place of loard Shri Krishna and RadhaRani. Mathura is located on the western bank of river Yamuna at latitude 27 degree 41 Minute N and 77 Degree and 41 Minuet E. Mathura city is located at a distance of 145 km south-east of Delhi and 58 km north-west of Agra in the state of Uttar Pradesh(India). We have chosen the area nearby Mathura refinery for collection of sample and some sites at a cooperatively large distance from refinery so that the effect of refinery can clearly be observed. Refinery is located along the Delhi-Agra National Highway about 154 KM away from Delhi. The major secondary processing units provided were Fluidised Catalytic Cracking Unit (FCCU), Vis-breaker Unit (VBU) and Bitumen Blowing Unit (BBU). The original technology for these units was sourced from erstwhile USSR, UOP etc. Soaker drum technology of EIL was implemented in VBU in the year 1993. Mathura Refinery is having its own captive power plant, which was augmented with the commissioning of three Gas Turbines (GT) and Heat Recovery Steam Generator (HRSG) in phases from 1997 to 2005 using Natural Gas(NG) as fuel to take care of environment.



3 EXPERIMENTAL DETAIL

In the present study radon–thoron concentration was measured by using the twin chamber dosimeter cups fitted with Solid State Nuclear Track Detectors (SSNTD). All dosimeters were fitted with 12µm thick, LR-115 type II peliculable, cellulose nitrate in the form of solid films. (Manufactured by Kodak Pathe, France) These dosimeter cups fitted with LR-115 type II films were placed in 20 rooms in different dwellings near the Refinery Area and some dwellings are chosen at a relatively large distance from refinery to compare the effect of refinery. The twin chamber dosimeter was developed at Bhabha Atomic Research Centre (BARC) [7]. Mumbai and is shown in Fig. 1. It consists of two cylindrical compartments each of a length of 4.1 cm and a radius of 3.1 cm with inner volume of 124 cm3, These dimension are chosen on the basis of ratio of effective volume of the cup and its total volume so that maximum tracks can be recorded. Out of the two compartment one compartment ‘M’ fitted with a piece of the detector film (SSNTD) of size 3 cm × 3 cm is used for the measurement of radon only, radon diffuses into it from the ambient air through a semi-permeable membrane of 25µm thickness having diffusion coefficient in the range of 10-8 to 10-7 cm-2 s-1[8]. It is reaches the steady state concentration inside the compartment in about 4.5 Hrs. It allows to buildup of about 90% of radon gas in the compartment and prevents the development of thoron gas concentration by more than 99% [7], [9]. Other compartment F fitted with another piece of detector film allows both radon and thoron gases to diffuse in it through a glass fiber filter paper of thickness of 0.56 mm and hence the tracks developed on second piece of the detector film placed in this chamber are related to the concentrations of both the gases. All these detectors were exposed in 20 dwellings nearby refinery area of Mathura city for a period of 90 days. After exposure the detector films were taken out of the cup and then etched in 2.5N NaOH solution at 600 ^{0}C for a period of about 90 minute inside a constant temperature water bath for removing a bulk thickness of 4µm, this leaves a residual detector for thickness of 8µm and then the films were pre-sparked using a spark counter [10] operating at a high voltage of about 900 V to completely develop the track holes which was not developed in the etching process. After that track density at each detector film was calculated by counting the tracks developed on the detector films, for which spark counter was used at a voltage corresponding to the plateau region of the counter (450 V). Thus the concentrations of radon were calculated from the observed track densities using appropriate calibration factors. From track density radon (CR) and thoron (CT) concentration were calculated using the

sensitivity factor determined from the controlled experiments [7], [11].

$$C_R(Bqm^{-3}) = \frac{T_m}{d \times S_m} \quad (1)$$

$$C_T(Bqm^{-3}) = \frac{T_f - d \times C_R \times S_{rf}}{d \times S_{tf}} \quad (2)$$

Where, C_R is the radon concentration, C_T is thoron concentration, Tm is track density in Membrane compartment, T_f is track density in filter compartment, d is exposure time. Sensitivity factor for membrane compartment (S_m)= 0.019 ± 0.003 Tr $cm^{-2}d^{-1}/Bqm^{-3}$ Sensitivity factor for radon in filter compartments (S_{rf})= 0.020 ± 0.004 Trc m^{-2} d^{-1}/Bq m^{-3} Sensitivity factor for thoron in filter compartment (S_{tf})= 0.016 ± 0.005 $Trcm^{-2}d^{-1}/Bqm^{-3}$.

The inhalation dose (D) in mSv y-1 was estimated using the relation [7].

$$D = \{0.17 + 9F_R)C_R + (0.11 + 32F_T)C_T\} \times 7000 \times 10^{-6} \quad (3)$$

Where F_R and $F_T$are equilibrium factor for radon and thoron respectively. The values are taken as 0.4 and 0.1 for radon and thoron [12] .

4 RESULTS AND DISCUSSION

The results of the measured radon/thoron concentration and inhalation for the study area are shown in table-1 given below indicate that radon concentrations vary from 35.1 ± 4.5Bq m-3 to 84.8 ± 7.0Bq m-3 with an average value 57.1Bqm-3. Here we observe that the average value of radon concentration (57.1Bq/m3) is found to be little greater than the average value of 40 Bqm–3, reported for the dwellings worldwide [13]. This may be due to little more concentration of radioactive content in soil and building materials due refinery waste in study area. Instead, in most of the buildings the radon concentration is below the level of concern i.e. 150Bqm-3 and none of the dwelling have value of radon concentration greater than the action level i.e. 200-600Bqm-3, which was recommended by ICRP [14]. The variations in the values of radon concentration in these building may be due to different ventilation conditions, different types of construction materials used. It has been also observe here that the dwelling having granite flooring have higher values of radon concentration (≥ 50Bqm-3) than that of having cemented flooring. The dwelling which is comparably close to the refinery also shows little higher values (≥70Bqm-3) of radon concentration which may be due to radium content from refinery waste. As expected Thoron and its daughter contributes a little towards the radiation dose. It can be seen from the table that thoron concentrations vary from 3.5 ± 0.3Bq $m^{-3}$to 24.3 ± 2.2Bq m^{-3} with an average value of 7.4Bq m^{-3}. It can be seen that the concentration of thoron are also high in the dwelling situated close to refinery. The annual effective dose received by the resident of study area is found to vary from 1.0mSv y^{-1} to 2.4mSv $y^{-1}$with an average value of 1.6mSv y^{-1} which is also less than the lower limit of action level (3mSvy^{-1}) recommended by International Commission on Radiological Protection (ICRP). Table 2 shows the comparison of radon concentration levels with some other studies of the area in different states of India and Table 3 shows the comparison of thoron concentration levels with some other studies of different countries. There is not much variation in radon/thoron concentrations except in the houses very nearby to the refinery which shows higher values. These may be due to that there are various toxic gases which are released by refinery in the atmosphere.

5 CONCLUSION

House wise analyses of radon, thoron and their daughter's concentrations (Bq m^{-3}) shows some variation in all the houses so we may conclude here that the radon and thoron concentration inside the dwellings is greatly affected by their exhalation and ventilation parameters. The variation the concentration level of radon and thoron may also depends on the materials used to construct the building and also here some effects has been noticed due to oil refinery. Although these effects due to refinery are not so much and are under the recommended levels. Also this variation in the concentration of radon, thoron and their daughters may be attributed to the variation in primordial radioactivity in the soil and building materials. Finally it was concluded from the study that the inhalation dose, radon and thoron concentrations are within the permissible limits.

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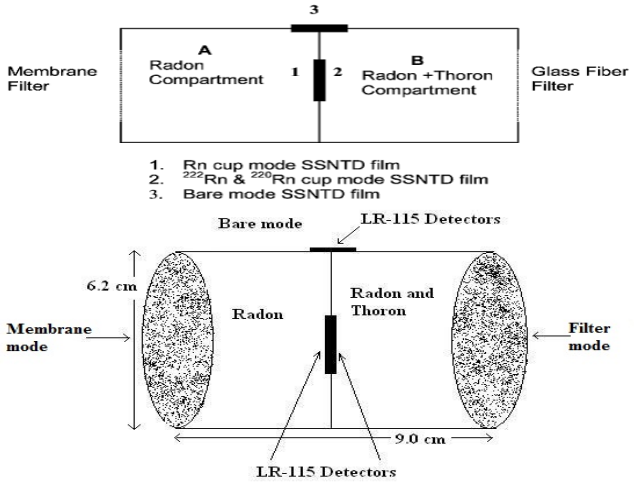

***FIG. 1** RADON- THORON TWIN CHAMBER DOSIMETER CUP SYSTEM*

***TABLE-1**: RADON AND THORON CONCENTRATION AND INHALATION DOSE IN DWELLINGS OF MATHURA CITY OF UTTAR PRADESH STATE OF INDIA*

Number of dwellings (Flooring/distance from Refinery)	C_R *(Bq m^{-3})*	C_T *(Bq m^{-3})*	*D (Inhalation dose) (mSv y^{-1})*
1 (Cemented)	*41.0±4.8*	*3.8±0.4*	*0.11*
2 (Granite)	*55.5±5.7*	*5.8±0.5*	*0.15*
3 (Granite)	*65.6±5.7*	*8.3±0.7*	*0.19*
4 (Granite)	*61.2±6.1*	*7.1±0.6*	*0.17*
5 (Cemented)	*48.1±5.2*	*4.5±0.4*	*0.13*

6 (Cemented)	*35.1±4.5*	*3.5±0.4*	*0.10*
7 (Cemented)	*40.1±4.8*	*3.6±0.4*	*0.11*
8 (Cemented)	*45.9±5.1*	*5.2±0.5*	*0.13*
9 (Cemented)	*53.8±5.6*	*6.8±0.6*	*0.14*
10 (Close to refinery)	*79.7±6.8*	*13.9±1.1*	*0.22*
11 (Granite)	*69.2±6.3*	*10.5±0.9*	*0.18*
12 (Granite)	*60.0±5.9*	*8.1±0.7*	*0.16*
13 (Cemented)	*52.0±5.5*	*5.4±0.5*	*0.14*
14 (Close to refinery)	*84.8±7.0*	*24.3±1.8*	*0.24*
15 (Granite)	*63.5±6.1*	*7.9±0.7*	*0.17*
16 (Cemented)	*44.6±5.1*	*4.6±0.5*	*0.12*
17 (Cemented)	*49.2±5.4*	*5.1±0.5*	*0.13*
18 (Close to refinery)	*79.0±6.9*	*7.7±0.6*	*0.21*
19 (Close to refinery)	*71.5±6.4*	*8.3±0.7*	*0.19*
20 (Cemented)	*43.5±5.0*	*4.0±0.4*	*0.11*
Minimum *Maximum* *Average*	*35.1±4.5* *84.8±7.0* *57.1*	*3.5±0.4* *24.3±1.8* *7.4*	*0.10* *0.24* *0.16*

Table 2: *Comparison of radon concentration levels with some other studies of the india.*

S. No	*IndoorRadon level(Bq/m^{-3})*	*Area*	*References*
1	*12-104*	*Faizabad (U.P.)*	*[15]*
2	*12-190*	*Jodhpur (Rajasthan)*	*[16]*
3	*15-218*	*Amritsar (Punjab)*	*[17]*
4	*13-143*	*All India*	*[18]*
5	*66-104*	*Northern Haryana*	*[6]*
6	*36-140*	*HamirpurandUna (HimachalPradesh)*	*[19]*
7	*18-62*	*Jaipur(Rajasthan)*	*[20]*
8	*35-84*	*Present Study*	

TABLE 3: *COMPARISON OF INDOOR RADON CONCENTRATION LEVELS WITH SOME OTHER STUDIES IN DIFFERENT COUNTRIES.*

S. No.	*Average indoor radon concentration (Bq m^{-3})*	*Region*	*References*
1.	*140*	*Czech Republic*	*[21]*
2.	*86*	*Spain*	*[21]*
3.	*123*	*Finland*	*[22]*
4.	*97*	*Austria*	*[23]*
5.	*48*	*Belgium*	*[24]*
6.	*68*	*France*	*[25]*
7.	*89*	*Ireland*	*[26]*
8.	*70*	*Italy*	*[27]*
9.	*115*	*Luxembourg*	*[28]*
10.	*112*	*Romania*	*[29]*
11.	*108*	*Sweden*	*[30]*
12.	*130*	*Turkey (Giresun)*	*[31]*
13.	*57*	*Present Study*	

7 REFERENCES:

[1] Baird, R.D., et al., 1990, Management and Disposal Alternatives for NORM Wastes in Oil Production and Gas Plant Equipment, prepared for American Petroleum Institute, Dallas, Texas

[2] K. P. Smith et al., "Assessment of the Disposal of Radioactive Petroleum Industry Waste in Nonhazardous Landfills Using Risk-Based Modeling," Environ. Sci. Technol. 37, 2060 (2003)

[3] Mullar Associates, Handbook of Radon in Buildings; detection, Safety and control, Hemisphere Publishing Corporation New York, USA (1988).

[4] K. P. Smith, "An Overview of Naturally Occurring Radioactive Materials (NORM) in the Petroleum Industry," Argonne National Laboratory, ANL/EAIS-7, December 1992

[5] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources Effects and Risks of Ionizing Radiation: Exposure from Natural Radiation Sources (1998) vol. I Annex-B. New York

[6] Chauhan R.P, (2010). Monitoring of radon, thoron and their progeny in dwellings of Haryana. Indian J of Pure & App Physics, 48: 470-472.

[7] Mayya Y. S., Eappen, K. P., and Nambi, K. .S V., Methodology for mixed field inhalation dosimetry in monazite areas using a twin-cup dosimeter with three track detectors, Radiation Protection Dosimetry, 77(3), 177–184 (1998).

[8] Ramachandran T. V. and Subba Ramu M. C., Variation of equilibrium factor F between radon and its short decay products in an indoor atmosphere, Nuclear Geophysics, 8, 499–503 (1994).

[9] Jojo P.J., Study of Radon and its progeny using etched track detectors and micro analysis of uranium, Ph. D Thesis, Aligarh Muslim University, Aligarh, 1993, P. 123.

[10] Nikolaev VA, Ilic R (1999) Etched track radiometers in radon measurements: a review Radiat Meas 30: 1-13.

[11] Dwivedi K.K., Mishra R., Tripathy S.P., A. Kulshreshtha, Singh D., Srivastava A., Deka P., Bhattarcharjee B, Ramachandran T.V., Nambi K.S.V. – Simultaneous determination of radon, thoron and their progeny in dwellings, Radiation Measurement, 2001, 33,7-11.

[12] UNSCEAR, United Nation Scientific Committee on the Effects of Atomic Radiation, Sources and Effects of Ionizing Radiation, United Nations: New York (1999).

[13] UNSCEAR, United Nation Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation, United Nations: New York (2000).

[14] ICRP, International Commission on Radiological Protection, ICRP Publication No. 65 Pergamon Press, Oxford (1993)

[15] Verma Deepak and Khan M. Shakir, Measurements of indoor Radon and Thoron in the Dwellings of Faizabad City Using Plastic Track Detectors, Indian J Pure & Applied Physics, 5, 219–222(2013).

[16] Kumar S., Gopalani Deepak, Ramaseshu P. and Nagaratnam A. Estimation of indoor radon levels in cities of Rajasthan by SSNTD. Radiation Protection Dosimetry, 37(2), 127–131 (1991).

[17] Virk H. S., and Sharma Navjeet, Indoor radon levels and inhalation doses to population in Punjab, Current Science, 78(12), 1418–1420 (2000).

[18] Ramachandran T. V., Indoor radon levels in India: current status of the coordinated nationwide study using passive detector technique, Proceedings of XIth National Symposium on SSNTD\ Amritsar, 50 (1998).
[19] Virk H. S. and Sharma Navjeet, Indoor radon/thoron levels and inhalation dose to some population in himachal Pradesh, Indian J Environment Monitoring, 4, 162–165 (2002).
[20] Sharma Jyoti, Mahur A. K., Kumar Rupesh, Varshney Rati, et al., Comparative study of indoor radon, thoron with radon exhalation rate in soil samples in some historical places at Jaipur,Rajasthan, India. Advances in Applied Science Research, 3(2), 1085–1091 (2012).
[21] UNSCEAR, United Nation Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation, United Nations: New York (1993)
[22] Castren O., Radon reduction potential of Finnish dwellings. Radiation Protection Dosimetry, 56, 375–378 (1994).
[23] Friedmann H., Atzmullar C., Breitenhuber P. et al., The Austrian radon project. The science of the Total Environment, 272(1–3), 211–212 (2001).
[24] Zhu H.C., Charlet J M. And Poffijn A., Radon risk mapping in southern Belgium. The science of the Total Environment, 272 (1–3), 203–210 (2001).
[25] Baysson H., Billon S., Laurier D., Rogal A. and Tirmarche M., Seasonal correction for estimation radon exposure in France dwellings. Radiation Protection Dosimetry, 104(3), 245–252 (2003).
[26] Fennel S. G., Mackin G. M., Madden A. T. et al., Radon exposure in Ireland. Internatioanl Congress Series, 1225, 71–77 (2002).
[27] Bochicchio F., Campos Venuti G., Nuccetelli C. et al., Results of the representative Italian National survey on radon indoors. Health Physics, 71(5), 743–750 (1996).
[28] Kies A., Feider M., Biell A. and Rowlinson L., Investigation on the dynamics of indoor radon concentrations. Environmental International, 22(Supp. I), S805–808 (1996).
[29] Szacsvai K., Cosma C. and Cucos A., Indoor radon exposure in Cluj-Napoca city Romania. Romanian J of Physics 58, S273–S279 (2013).
[30] Swedjemark G.A. and Hubbard L.M., Challeges in comparing radon data sets: 1955–1990", INDOOR AIR '93. Proceedings of the 6th International Conference on Indoor Air Quality and Climate, Helsinki, Finland, July 4–8, Vol.4, 431–436 (1993).
[31] Celik N., Cevik U., & Kucukomeroglu B., Determination of indoor radon and soil radioactivity levels in Giresun, Turkey. Journal of Environmental Radioactivity, 99, 1349–1354 (2008).