Studies on indoor and outdoor radon (^{222}Rn) and its short-lived progeny concentration for the assessment of safety and health of public in the environment of coastal Kerala

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Introduction

Radon and its progeny produced by the decay of naturally occurring radioisotope Uranium (^{238}U) are ubiquitous in the environment. The radioactive gas along with its radioactive progeny enters into the lungs especially by means of breathing and delivers dose to the tissues. Breathing of air containing excessive concentration of radon can deliver high radiation dose to the tissues and can be hazardous. This adds to the probability of incidence of radiogenic lung cancer also.

About 50% of the total radiation dose received on an average by the Indian population is due to breathing of the radon and its progeny present in dwellings. Hence the recommendations by the International Commission on Radiological Protection (ICRP) together with the Atomic Energy Regulatory Board (AERB) have stressed the need for measurement of concentration of radon and its progeny especially in dwellings. The concentration of these radionuclides in the environment is not uniform throughout the world.

The method of measurement of radon and its progeny may be based on an active technique, which involves the pumping of indoor air containing the gases or its progeny into or through a detecting system (eg. Collection technique) or a passive technique where the concentrations are measured under natural conditions by exposing the detectors indoors (SSNTD technique). Collection technique is broadly classified as scintillation cell method and filter paper method.

The walls of the scintillation cell are coated with phosphor like ZnS activated with silver. The sample of air containing radon is taken in the cell. Alpha particle from the daughters fall on ZnS which produces light pulses. These light pulses fall on a photomultiplier which is coupled to the cell. The photomultiplier converts light pulses into electrical pulses. The activity of radionuclides is measured in terms of these pulses. The sensitivity of this method depends on length of the counting period and the background of the cell.

In filter paper method sample is drawn through a special filter paper (Millipore filter of type AA) at a convenient known flow rate using a low noise pump. The activity of the progeny collected on the filter paper is measured by counting the alpha particles, emanating from the sample collected with the help of the phosphor- photomultiplier coupling alpha counter. Computerized least squared program (Rangarajan C, 1976) analyses the decay data and gives the Ra-A, Ra-B and Ra-C concentrations in Bqm^{-3} and PAECs of radon progeny. The accuracy of this method is controlled by the counting statistics and the error can be less than 5% provided the count rate is sufficiently large.

Coastal Kerala known for its splendidly beautiful natural environment has become a major source of radioactive monazite. Due to the monazite placer deposit, regions of enhanced radiation level are identified along coastal Kerala and are termed as High Background Radiation Areas (HBRA). In this context, the studies on behaviour of the radionuclide with its progeny concentration in the environment have assumed great significance.

Extensive and systematic investigations in different environmental matrices have been carried out to obtain a clear understanding of the basic dynamics of radon and its progeny in the environment. Considering the serious hazards of environmental pollution and keeping in view of the present and future industrial activities in and around coastal

Kerala, systematic studies on radon and its progeny concentration in different environmental matrices have been undertaken.

Materials and methods

The application of nuclear techniques to study the radioactivity in the environment of a region requires precise detection and careful analysis of nuclear radiation. In present investigation, various materials and instruments were used for radon measurement in soil, air and groundwater. The measurement techniques are based on the detection of emissions from radioactive decay of radon and its daughter products. Most of the methods are based on the detection of alpha particles. Brief description of these materials and methods are given below.

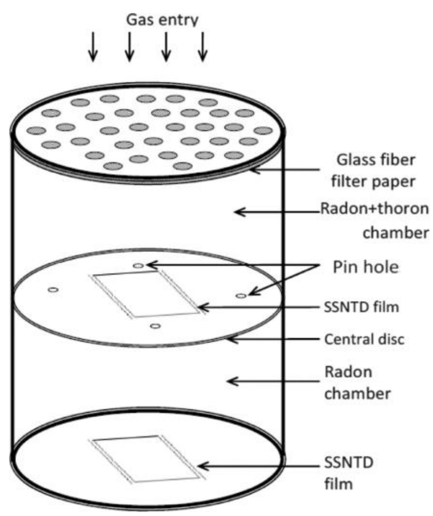
Solid State Nuclear Track Detectors

The alpha sensitive plastic track detector, LR-115 Type II has been used in present investigation for integrated measurement of radon. It is a 12μm thick film red dyed cellulose nitrate emulsion coated on inert polyester base of 100μm thickness and has maximum sensitivity for alpha particles, fission fragments and ionizing particles with high enough LET. The upper threshold energy in LR-115, which produces the tracks as through holes, is 4.8 MeV (Abu-Jarad et al., 1980). The film can be used to record the tracks of protons with energy <100 KeV. However, it is insensitive to X or γ- rays, photons, electrons and high energy protons. For fast neutrons, it has low detection efficiency (10^{-5} tracks/neutron) (Khan, 1975). The detection efficiency of alpha particles for the detector at normal incidence is about 50% for energies between 1.5 MeV and 4.8 MeV.

In Solid State Nuclear Track Detectors (SSNTD), thin plastic dielectric film of either cellulose nitrate (CN) or polycarbonates is exposed to the alpha radiation emitted by radon and its progeny and the tracks recorded by the film are read out. For indoor measurements, normally CN films are used as they are very specific to alpha radiation detection. In case of thin films tracks are counted after etching with the help of a spark counter. In case of thick films tracks are counted directly by using optical microscope. The SSNTD films can be exposed in three different modes to measure the concentrations of radon in Bqm^{-3} and PAEC for radon progeny in terms of WL units. In field either a bare film of SSNTD is exposed or a sample of air is allowed to diffuse into a suitable cup through either a filter paper or a membrane fixed over the cup.

Pin-hole based twin cup Radon-Thoron dosimeter

In the present study a new "pin-holes based twin cup dosimeter" has been utilized, with single face for entry of radon (^{222}Rn) and thoron (^{220}Rn) gases from environment. The design of such dosimeter was possible only because of a newly developed pin-holes based ^{222}Rn-^{220}Rn discrimination technique and the associated theory by selecting a suitable chamber volume and dimension of pin-holes, it is possible to cut off ^{220}Rn entry into the chamber volume and allow only ^{222}Rn (Sahoo et al., 2013). The new design of this dosimeter system has two compartments separated by a central pin-holes disc made up of HDPE material, acting as ^{220}Rn discriminator. Each chamber has a length of 4.1cm and radius of 3.1 cm (same dimensions as in the twin cup dosimeter developed by (Eappen & Mayya, 2004). The first compartment (named as 'radon + thoron' chamber) samples ambient air at the entry of which, the particulates are restricted by using a filter paper. The air containing radon and thoron from this compartment diffuses to the second compartment (named as “radon” chamber) through pin-holes, acting as a diffusion barrier that cuts off the entry of thoron into this chamber due to its short half-life (55.6 s). Hence, only radon enters into this compartment. The tracks registered in LR-115 placed in first and second chamber are corresponding to the radon + thoron and radon concentration in the atmosphere respectively. Schematic diagram of the pin-hole dosimeter has been shown in the Fig.



The schematic diagram of pin-hole based twin cup dosimeter representing radon and radon+thoron chambers.

Constant temperature bath

A constant temperature bath from Polltech Instruments, Mumbai is used for the etching of LR-115 film in order to develop the tracks in these detectors. This instrument has the temperature range from ambient to 95 ^{0}C with an accuracy of ± 0.2 ^{0}C. It consists high and low heat arrangements to heat up the sink water with fine and course temperature setting arrangement. It has solid state time proportional temperature controller with digital display arrangement and auto-cut system at set temperature. The main tank is a stainless steel interior fitted with a circulator for uniformity of temperature in externally connected circuit. The double walled beakers with chemical solution for etching of the plastic detectors can be connected with the circulator through rubber pipes.

Spark counter

The spark counting technique, which is the most successful and widely used technique for counting etched tracks in plastic track detectors was first invented by Cross and Tommasino (1970) and has been developed and discussed in a number of publications (Somogyi et al., 1978; Azimi-Garakani et al., 1981; Tommasino et al., 1986, Durrani and Bull, 1987). When the plastic track detector is chemically etched, the through holes are produced along the tracks. Spark counter is an instrument used to count the number of tracks on the films. The thin etched detector, which is an insulating material, is placed between two electrodes of the spark counter forming a capacitor and covered with an aluminized plastic foil (a very thin layer of aluminium evaporated onto a mylar backing). The aluminized side of the plastic foil is in contact with the thin detector. The thick conductive electrodes of a spark counter are commonly made of brass. It is sometime necessary to put a relatively heavy weight on top of the plastic foil to have an intimate contact between the thin detector and the electrodes. With application of a high voltage across the capacitor C, an electrical discharge or spark takes place through a track-hole. The voltage pulse produced across the resistor, R, can easily be counted electronically by a scalar (Azimi-Garakani et al., 1981).

The spark passing through a track hole has enough energy to evaporate the thin layer of aluminium coating and produces a much larger hole in the aluminium electrode. Because of the evaporation of the aluminium there exist a short circuit in the electrode, and hence second spark cannot occur in the same track hole. As a result the spark is stopped, the capacitor C is charged again. Consequently, the spark shifts randomly from one track hole to another until all track holes are counted. The evaporated spot on the aluminium, which have the diameter of about 100 μm are equal to the number of sparks and hence to the number of track holes in the plastic track detector. The aluminium replica can easily be counted by an optical microscope or a microfiche reader.

Results and Discussion

The pelliculable LR-115 type-II plastic track detectors (SSNTDs) of size 3 cm × 3 cm have been deployed in the pin-holes based twin cup dosimeters for indoor radon, thoron and their progeny measurements simultaneously in 20 dwellings of Kannur district, Kerala from 1st October 2014 to 31st September 2015. To study seasonal variation pattern of indoor emanation, the whole year has been divided into three sessions of four months each covers post monsoon, pre-monsoon and monsoon. The dwellings in each village were selected on the bases of building material like mud, bricks and concrete, flooring materials like granite, ceramic tiles, cement and red oxide and roofing materials like concrete, wood, asbestos and roof tiles.

The dosimeters were installed in the dwellings for a period of about 90 days in such a way that they are atleast 1 m above the ground level and 15 cm away from the nearest wall or roof. In order to study the seasonal variation of radon and thoron levels, the dosimeters were reloaded and re-installed after every 4 months. After the exposure time, the detector films were removed, etched with 2.5 N NaOH solution for 90 minutes in a constant temperature bath at 60^{0}C. The detector films are then washed in distilled water. After washing, the detectors are air dried in dust free environment. The sensitive layer of the detectors are peeled off from their plastic bases and the track density was obtained by using the standard spark counter (model PSI-SC1) with operating voltage (750V) as well as the pre-sparking voltage (900V).

The radon concentration C_r and thoron concentration C_t in the filter compartment and the pinholes compartment respectively was calculated by using formulae given as:

C_r (Radon conc.) = $T_1/(d.k_r)$

C_t (Thoron conc.) = $(T_2 - d.C_r k_r)/(d.k_t)$

Where T_1(M) and T_2(F) represents track densities of radon in membrane mode and radon+thoron in filter mode respectively, d represents the number of days exposed, k_r (0.0170 ± 0.002 tr. Cm^{-2} per Bq.d.m^{-3}) and k_t (0.010 ± 0.001 tr. Cm^{-2} per Bq.d.m^{-3}) are the calibration factors of radon and thoron in 'radon + thoron' compartment (Sahoo et al., 2013).

Annual dose received by the inhabitants in the dwellings under study in mSv was estimated using the relation given by Sannappa et al., 2003 & Mayya et al., 1998.

$D = [(0.17+9F_r)C_r + (0.11+32F_t)C_t] \times 7000 \times 10^{-6}$ mSv.

Where, F_r and F_t are the equilibrium factors for radon and thoron respectively. F_r = 0.4 and F_t = 0.1 (UNSCEAR 1992).

The all 20 dwellings were divided into 8 categories based on their building materials of floor and roof. They are discussed in the tables below.

Table1. The annual effective dose for pre monsoon season

Types of house	**Samples**	**Pre monsoon**				**Annual effective dose**
		T1(M) Tr/cm^2	**T2(F) Tr/cm^2**	**C_r Bq/m^3**	**C_t Bq/m^3**	**D mSv**
Granite floor and concrete roof	S1	430	485	238.9	61.1	7.72
	S2	396	453	220	63.3	7.27
	S3	415	470	230.5	61.2	7.50
	Avg.	**413.7**	**469.3**	**229.8**	**61.9**	**7.49**
Ceramic tile floor and concrete roof	S4	390	443	216.6	59	7.08
	S5	398	452	221.1	60.2	7.22
	Avg.	**394**	**447.5**	**218.9**	**59.6**	**7.15**
Cement floor and rooftile roof	S6	84	108	46.7	26.6	1.84
	S7	93	112	51.7	21	1.85
	Avg.	**88.5**	**110**	**49.2**	**23.8**	**1.84**
Cement floor and asbestos roof	S8	296	342	164.4	51.2	5.52
	S9	287	336	159.4	54.5	5.46
	S10	271	321	150.6	55.5	5.26
	Avg.	**284.7**	**333**	**158.1**	**53.7**	**5.41**
Cement floor and concrete roof	S11	250	295	138.9	49.9	4.82
	S12	231	277	128.3	51.2	4.57
	S13	228	272	126.7	48.8	4.47
	Avg.	**236.3**	**281.3**	**131.3**	**49.9**	**4.62**
Red oxide floor and wood+ rooftile roof	S14	55	73	30.6	19.9	1.26
	S15	64	83	35.6	21	1.42
	Avg.	**59.5**	**78**	**33.1**	**20.5**	**1.34**
Cement floor and asbestos+ rooftile roof	S16	199	240	110.6	45.5	3.97
	S17	184	223	102.2	43.4	3.70
	S18	175	212	97.2	41.2	3.51
	Avg.	**186**	**225**	**103.3**	**43.4**	**3.72**
Red oxide floor and wood+ asbestos +rooftile roof	S19	117	147	65	33.3	2.48
	S20	91	119	50.6	31	2.05
	Avg.	**104**	**133**	**57.8**	**32.2**	**2.26**
Average values for the pre monsoon season				**129.3**	**44.9**	**4.45**

Table2. The annual effective dose for monsoon season

Types of house	Samples	Monsoon				Annual effective dose
		T1(M) Tr/cm^2	T2(F) Tr/cm^2	C_r Bq/m^3	C_t Bq/m^3	D mSv
Granite floor and concrete roof	S1	502	574	278.9	79.9	9.21
	S2	464	538	257.8	82.2	8.70
	S3	470	541	261.1	78.9	8.71
	Avg.	**478.7**	**551**	**265.9**	**80.3**	**8.87**
Ceramic tile floor and concrete roof	S4	465	535	258.3	77.8	8.61
	S5	472	540	262.2	75.6	8.67
	Avg.	**468.5**	**537.5**	**260.3**	**76.7**	**8.64**
Cement floor and rooftile roof	S6	112	145	62.2	36.7	2.49
	S7	108	133	60	27.8	2.22
	Avg.	**110**	**139**	**61.1**	**32.3**	**2.35**
Cement floor and asbestos roof	S8	350	412	194.4	69	6.72
	S9	363	429	201.7	73.3	7.02
	S10	342	399	190	63.3	6.48
	Avg.	**351.7**	**413.3**	**195.4**	**68.5**	**6.74**
Cement floor and concrete roof	S11	301	356	167.2	61.2	5.83
	S12	268	327	148.9	65.5	5.44
	S13	281	343	156.1	68.9	5.71
	Avg.	**283.3**	**342**	**157.4**	**65.2**	**5.66**
Red oxide floor and wood+rooftile roof	S14	70	92	38.9	24.4	1.59
	S15	86	106	47.8	22.2	1.77
	Avg.	**78**	**99**	**43.4**	**23.3**	**1.68**
Cement floor and asbestos+rooftile roof	S16	254	298	141.1	48.9	4.85
	S17	205	254	113.9	54.4	4.26
	S18	241	285	133.9	48.9	4.66
	Avg.	**233.3**	**279**	**129.6**	**50.7**	**4.59**
Red oxide floor and wood+asbestos+rooftile roof	S19	124	163	68.9	43.3	2.82
	S20	101	135	56.1	37.8	2.35
	Avg.	**112.5**	**149**	**62.5**	**40.6**	**2.58**
Average values for the monsoon season				**155**	**57**	**5.41**

Table3. The annual effective dose for post monsoon season

Types of house	**Samples**	**Post monsoon**				**Annual effective dose**
		T1(M) Tr/cm^2	**T2(F) Tr/cm^2**	**C_r Bq/m^3**	**C_t Bq/m^3**	**D mSv**
Granite floor and concrete roof	S1	526	604	292.2	86.7	9.7
	S2	490	570	272.2	88.9	7.4
	S3	479	545	266.1	73.3	8.7
	Avg.	**498.3**	**573**	**276.8**	**82.9**	**8.6**
Ceramic tile floor and concrete roof	S4	467	539	259.4	80.1	8.7
	S5	445	518	247.2	81.2	8.4
	Avg.	**456**	**528.5**	**253.3**	**80.7**	**8.5**
Cement floor and rooftile roof	S6	130	164	72.2	37.8	2.8
	S7	123	150	68.3	30.1	2.5
	Avg.	**126.5**	**157**	**70.3**	**33.9**	**2.7**
Cement floor and asbestos roof	S8	378	445	210	74.4	7.3
	S9	400	463	222.2	70.1	7.5
	S10	354	417	196.7	69.9	6.8
	Avg.	**377.3**	**441.7**	**209.6**	**71.5**	**7.2**
Cement floor and concrete roof	S11	317	378	176.1	67.8	6.2
	S12	244	306	135.6	68.8	5.2
	S13	294	358	163.3	71.2	6.0
	Avg.	**285**	**347.3**	**158.3**	**69.3**	**5.8**
Red oxide floor and wood+rooftile roof	S14	75	101	41.7	28.8	1.8
	S15	98	116	54.4	20.1	1.9
	Avg.	**86.5**	**108.5**	**48.1**	**24.5**	**1.85**
Cement floor and asbestos+rooftile roof	S16	283	330	157.2	52.3	5.4
	S17	224	275	124.4	56.8	4.6
	S18	257	302	142.8	49.9	4.9
	Avg.	**254.7**	**302.3**	**141.5**	**53.0**	**4.9**
Red oxide floor and wood+asbestos+rooftile roof	S19	134	175	74.4	45.6	3.0
	S20	105	143	58.3	42.3	2.5
	Avg.	**119.5**	**159**	**66.4**	**43.9**	**2.75**
Average values for the post monsoon season				**161.7**	**59.8**	**5.57**

The work presented here emphasizes the long term measurements of radon and thoron concentrations in 20 dwellings using solid state nuclear track detectors. The houses chosen for installing dosimeters are based mainly on their building materials used to construct roof and floor.

The above tables show the seasonal variation of indoor radon and thoron concentrations in different types of dwellings in Kannur district of Kerala. In the present study it is observed that the minimum indoor radon concentration (33.1 Bq/m^3) was observed in the house which is made up of red oxide floor and wood+rooftile roof during pre monsoon season while the highest concentration (276.8 Bq/m^3) was recorded in the house of granite floor and concrete roof in post monsoon season. The average value of indoor radon concentration in the regions were calculated as 129.3 Bq m^{-3}, 155 Bq m^{-3} and 161.7 Bq m^{-3} for pre monsoon, monsoon and post monsoon respectively. The overall average value of indoor radon concentration was found 148.7 Bq/m^3 for the complete year. The minimum indoor thoron concentration (20.5 Bq/m^3) was recorded in the house which is made up of red oxide floor and wood+rooftile roof during pre monsoon season while the highest concentration (82.9 Bq/m^3) was recorded in the house of granite floor and concrete roof in post monsoon season. The average value of indoor thoron concentration in the regions were calculated as 44.9 Bq m^{-3}, 57 Bq m^{-3} and 59.8 Bq m^{-3} for pre monsoon, monsoon and post monsoon respectively. The overall average value of indoor thoron concentration in study area was found 53.9 Bq/m^3 for the complete year.

The average annual effective doses were calculated as 4.4 mSv, 5.4 mSv and 5.6 mSv for post monsoon, monsoon and pre monsoon respectively. The annual effective dose received by the inhabitants due to radon and thoron in the dwellings under study area varied from 1.26 to 9.7 mSv with an average value of 5.1 mSv for the complete year. The concentration of radon and thoron was observed less in pre monsoon and more in post monsoon season. This is because, in summer season, the houses remain open for long time which contributes in increasing air exchange rate, while in winter the rooms of the houses remain closed for long hours and thereby decreasing the chances of air exchanges. The measurement indicates normal levels of radon concentration and are within the safe limit 200 Bq/m^3 recommended by ICRP (1993) for dwellings. The annual dose was also well within the limit of 1-20 mSv for dwellings recommended by ICRP 2006. The data established from the present work will facilitates a scientific assessment, in future, of the impact of major and minor industrial activities in and around the region of coastal Kerala.

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