Universal Journal of Environmental Research and Technology
Available Online at: www.environmentaljournal.org

2011 Vol 1 7-21

Open Access

Review Article

^{220}Rn in indoor Environment of India: A Review

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Abstract

Data for ^{220}Rn in indoors and workplace environment is scare due to the general perception that its levels are negligible due to its shorter half life, and subsequently its contribution to the total inhalation dose is ignored, in the presence of other significant sources of natural radiation. The Bhabha Atomic Research Center (BARC), Mumbai, India has completed a countrywide monitoring program of ^{220}Rn along with ^{222}Rn in the dwellings using ^{222}Rn/^{220}Rn discriminating Solid State Nuclear Track Detector (SSNTD) based dosimeter systems with large participation of research groups from different parts of the country. Details about measurement, standardization of dosimeters and evaluation of the inhalation dose is presented in this paper. Results are compared with the values reported in literature for dwellings as well as in high background radiation areas.

Keywords: Dose, Indoor pollution, Inhalation, SSNTD, Thoron

1. Introduction

Topic on background radiation has evoked concern between scientist and layman alike in recent years due to the shift in focus of health effects from exposure of radiation from acute high to chronic low level. Globally many locations have higher levels of natural background radiation due to elevated levels of primordial radionuclides in the soil and their decay products like ^{222}Rn and ^{220}Rn in the environment. Of late, technologically enhanced naturally occurring radioactive material has also contributed to the burden of background radiation. It is estimated that inhalation of ^{222}Rn, ^{220}Rn and their short lived progenies contribute more than 54 % of the total natural background radiation dose received by the general population. Due to this it was necessary to supplement the external component with inhalation component. This component is not adequately estimated for any country so far on a national level. ^{220}Rn problem will also be a problem in industries which uses thorium nitrate. Including India, lamps using thoriated gas mantles are being still used for indoor and outdoor lighting and hawkers in rural as well as urban areas. Considering the fact that large amount of thorium nitrate is being handled by these industries, contribution to the inhalation dose of workers from ^{220}Rn gas emanated and build up of the progeny in ambient air may also be quite significant. It is not true from the recent studies showing high ^{220}Rn levels in living environments and work places across various countries and it is increasingly felt that it may be necessary to have data on ^{220}Rn in environment for obtaining a complete picture of inhalation dose. In this paper current status of ^{220}Rn levels in the indoor environment and workplaces as well as in other industries where large amount of ^{232}Th is being handled is being summarized. Methods of measurement and reported levels in literature are also summarized.

2. Brief Literature Survey on ^{220}Rn

The studies on uranium miners have established the presence of a positive risk coefficient for the occurrence of lung cancer in miners exposed to elevated levels of ^{222}Rn and its progeny, simultaneously there was a great upsurge of interest in the measurement of ^{222}Rn in the environment. It was also hoped that in conjunction with epidemiological studies, a large-scale ^{222}Rn surveys might lead to a quantitative understanding of the low dose effects of ^{222}Rn exposures. Considerable data is generated on the levels of ^{222}Rn in the environment (UNSCEAR, 2000). In contrast, data on ^{220}Rn is scare due to the general perception that its level is negligible due to its shorter half life (55 sec) and its contribution to inhalation dose is ignored, in the presence of other more significant natural radiation. This may not be true from the recent studies resulted in the observation of high ^{220}Rn in the living environments in various countries and is increasingly felt that it may be necessary to have information on ^{220}Rn levels in the environment for obtaining a complete picture of inhalation dose (Steinhausler *et al.,*1994; Porstendorfer, 1994).

^{220}Rn is discovered in 1899 by Owen's at McGill University in collaboration with Ernest Rutherford. Most of the early work focused on the fundamental physical properties of natural radioactivity, but some of it is still relevant to modern environmental consideration. Important step in ^{220}Rn research occurred in the atmospheric sciences when it was realized that ^{220}Rn and its progeny are a major source of atmospheric ions near the earth's surface, which are important to a wide range of atmospheric processes, including nucleation of water drops which are necessary for ^{222}Rn and formation of thunderstorms. ^{220}Rn and its progeny have been used as tracer in studies of atmospheric transport processes, such as eddy diffusion. Much of the early atmospheric research was by Israel and others (Israel *et al.,* 1968; Dolezalek, 1972) and the field has continued to be very active (Burchfield *et al.,* 1983). There are few industrial situations where ^{220}Rn can be found to be more in isolation from ^{222}Rn. Most of these are connected with industrial applications of ^{232}Th. ^{232}Th is a component in certain alloys, like magnesium, ply a small role in nuclear fuel cycle industry. But, the health problems associated with these applications have not been striking. Mining of ^{232}Th ore is done in well ventilated open pits. Wastes and tailings from ^{232}Th bearing ores processed for metals other than ^{232}Th can potentially release significant ^{220}Rn.

3. Review of Physical Properties of ^{220}Rn

^{232}Th is the ultimate progenitor of ^{220}Rn, its distribution in the earth's crust is important for controlling the production of ^{220}Rn. Tracers of ^{232}Th permeate almost all soils and rocks, in part due to the influence of ground water from which ^{232}Th can precipitate over geological time scales. ^{232}Th usually exist in plus four valence state. It is not highly soluble itself, but forms complex ions which are more soluble (Langmuir and Herman, 1980). ^{232}Th can be leached from primary source rock under proper conditions of acidity (pH) and oxidation potential (Eh). It then can be carried by water to other locations where it is in solution. Even though ^{232}Th is not as soluble as ^{238}U, there is some similarity in their geochemistry and soils enhanced in ^{238}U are often enhanced in ^{232}Th. In magmas and hard rocks, there is likely to be an even stronger correlation between ^{232}Th and ^{238}U deposits since here the respective geochemical processes have a greater similarity yet. The average concentration of ^{232}Th in soil is estimated to be 25 Bq kg^{-1} (UNSCEAR, 2000), with organic matter, there is some tendency with unusually high ^{232}Th content including monazite, thorite, zircon, sphene, and allanite. Rocks composed of granite or black shale are likely to have high ^{232}Th content. Monazite and zircon sands have an especially high concentration of ^{232}Th. In contrast, basalt, lime stone, and sand stone typically have a below average concentration. ^{232}Th is widely distributed in nature with an average concentration of 10 ppm in earth's crust in many phosphates, silicates, carbonates and oxide minerals. Natural ^{232}Th is present as nearly 100 % ^{232}Th isotope. In general ^{232}Th occurs in association with ^{238}U and rare earth element (REE) in diverse rock types; as veins of thorite, thorianite, uranothorite and as monazite in granites, syenites, pegmatite's and other acidic intrusions. Monazite also present in quartz-pebble conglomerate sand stones and in fluviatile and beach placers. In addition ^{232}Th is also found as an associate element with REE bearing bastnaesite in carbonates. Current knowledge of ^{232}Th resources in the world is limited due to the low-key exploration efforts arising out of insignificant demand. Apart from its main use in nuclear energy, as fertile material, ^{232}Th finds limited application in non nuclear areas, mainly as thorium nitrate for gas mantle industries and to a very limited extend as thorium oxide refractory, catalyst (for synthesis of either methane or mixtures of saturated and unsaturated hydrocarbons from mixtures of CO and H^2), throated tungsten wielding rods and in magnesium-based alloys. All these applications give raise higher inhalation dose rate to the public. The decay scheme of the ^{232}Th series is given in Table 1. Number of locations with higher content of ^{232}Th has been identified. Best known, perhaps are the monazite sands along the southern coast of Brazil, in Sri Lanka (Ceylon), and on the south tip of India. In the United States, the Triassic Conway granite of North Hampshire and coastal area of the southeast have large deposits of ^{232}Th. In contrast, ^{232}Th content of the oceans far from freshwater discharge is typically quite low, < 10^{-4} Bq kg^{-1} (Wedepohl 1978). ^{232}Th content in soil around this high background areas varied from 0.5 to 1000 Bq kg^{-1} (UNSCEAR, 2006). Jibri and Biere (2011) have reported that the natural radionuclides in the soil samples across the three farms varied from 2550.4±154.6 to 3208.9±188.7 Bq kg^{-1} for ^{40}K, 33.1±11.9 to 39.9±9.3 Bq kg^{-1} for ^{226}Ra, while for ^{232}Th it varied between 51.98±8.4 and 56.08±17.51 Bq kg^{-1} for the environment of Nigeria.

Table 1: Principal members of the ^{232}Th series (UNSCEAR, 1988)

Nuclide	Half-life	Major radiations and accompanying decay		
		Alpha (MeV)	Beta (MeV)	Gamma (MeV) and X rays (KeV)
^{232}Th	1.4×10^{10} y	3.95, 4.01	-	L
^{228}Ra	5.8 y	-	39 KeV	-
^{228}Ac	6.13 h	-	1170, 1740	L, 338, 911,969
^{228}Th	1.91 h	5.34, 5.42	-	L
^{224}Ra	3.62 d	5.69	-	241
^{220}Rn	55.6 s	6.29	-	-
^{216}Po	0.15 s	6.78	-	-
^{212}Pb	10.6 h	6.05	334, 573	238.6, 300.1
^{212}Bi	60.6 m	6.09	1520, 2250	L, 727,785,1620
^{212}Po	300×10^{-6} s	8.78	-	-
^{208}Tl	3.05 m	-	1280, 1520, 1790	L, 511,583,860,2614
^{208}Pb	Stable	-	-	-

Table 2: Properties of ^{220}Rn (UNSCEAR, 1988)

Boiling point	- 61.8 ^{o}C
Melting point	- 71 ^{o}C
Solubility in water:	
At 0 ^{o}C	0.51
20 ^{o}C	0.25
50 ^{o}C	0.14
Solubility in Acetone	8.0 at 0 ^{o}C
Diffusion Coefficient in air	0.1 cm^2 s^{-1} at STP
Diffusion Coefficient in water	1.1×10^{-5} cm^2 s^{-1} at 18 ^{o}C

The basic physical properties of ^{220}Rn are given in Table 2. Immediate parent of ^{232}Th is ^{224}Ra. This isotope is not always in equilibrium with ^{232}Th, particularly in ground water, in broad terms its concentration in soils and rocks will correlate well with ^{232}Th. Upon decay ^{224}Ra, the ^{220}Rn atom will experience recoil. If decay takes place within a mineral, the recoil range is of the order of 30 nm. So, ^{220}Rn atoms might be expected to remain trapped in the grains for the short time they exist before decay. A typical value for ^{220}Rn levels in the pore air of deep soil is estimated to about 20000 Bqm^{-3} (corresponding to a soil with about 25 Bqkg^{-1} ^{232}Th, porosity 50 %, density 1.5 gcm^{-3} and an emanation coefficient of 0.3). The typical values of ^{232}Th content and ^{220}Rn flux density in different materials are given in Table 3. Total known world reserve of ^{232}Th in reasonably assured reserves (RAR) and estimated additional reserve (EAR) categories are in the range of 2.23 MT and 2.13 MT respectively (IAEA, 1994) and is given in Table 4.

Table 3: Common values of ^{232}Th content and ^{220}Rn flux density (Shery, 1997)

Material	^{232}Th (Bq kg^{-1})	Flux density (Bq m^{-2} s^{-1})
Soil	10 70	0.5 – 5
Limestone	5	0.04
Punic stone (thick)	100	0.5
Black shale	Up to 400	-
Granite	100 – 200	-
Sandstone	5	0.05
Basalts	2 – 15	-
Concrete	25	0.04
Gypsum	10	0.1
Monazite sand	4×10^4 to 3×10^5	-

Table 4: Estimated ^{232}Th reserves (Tons of Thorium metal) (Schery, 1992)

Country	RAR	EAR
Australia	19000	---
Brazil	606000	700000
Canada	45000	128000
Greenland	54000	32000
Egypt	15000	309000
India	319000	---
Norway	132000	132000
South Africa	18000	---
Turkey	380000	500000
United States	137000	295000

In the RAR category, the deposits in Brazil, Turkey and India are in the range of 0.60, 0.38 and 0.32 million tons respectively. ^{232}Th deposits in India has been recently reported to be in the range of 0.65 MTs. Large known reserve of ^{232}Th are contained in the beach sand and inland placer deposits of monazite, a mixture of phosphate minerals with chemical formula (RE/TH/U) PO_4. Monazite is a primary source of light REE and ^{232}Th and a secondary source of phosphate and uranium. Hazards from ^{232}Th can be from both external and internal sources. External hazards are due to high energy beta and gamma rays, while the internal hazards are due to alpha emitting nuclides deposited inside the body. Internal hazards are mainly by way of inhalation of ^{232}Th bearing dust and short-lived decay products of ^{220}Rn gas. Typical activity content of beach sands and monazite of Indian continent is given in Table5.

Table 5: Typical radioactivity content of beach sands and monazite of Indian continent (IAEA, 1994)

Material	Activity concentration (Bq kg^{-1})		Radiation field(μGyh^{-1})
	^{232}Th	^{238}U	
Raw sand	0.32 – 6.44	0.04- 0.74	00.5 – 5.0
Monazite	322	37	180 - 250

Radon isotopes are inert gases which form chemical compounds only with difficulty (Stein, 1987). ^{220}Rn progeny in decay chain up to ^{208}Tl are the most for airborne dosimetry, particularly the alpha – particle emitters. All progenies are chemically reactive metals which readily oxidize and attach to surfaces like walls or the surface of aerosols. Immediately after decay the recoiling nucleus of these progeny is most frequently in a positive charge state. If unattached to aerosols, these, usually existing as molecular clusters have a diffusion coefficient in air about 0.05 cm^2 s^{-1}, with the exact value depending on the properties of the air like moisture content and the presence of trace gases (Porstendorfer, 1994). Existence of high ^{220}Rn levels were already investigated thoroughly where ever surveys were carried out with the ^{222}Rn - ^{220}Rn discriminative measurements, behavior of the ^{220}Rn and its progenies and their effects on human health have not been clearly elucidated yet. ^{220}Rn can migrate to earth's atmosphere exhibit with indoors and outdoors, can be inhaled mainly its progeny, through inhalation. ^{220}Rn is a natural production of ^{232}Th series in the earth's crust like soil, rocks and also in building materials (Schery and Wasiolek, 1997). Estimates show a range of values for ^{232}Th levels in the ground surface. As a result, UNSCEAR (2000) estimated a world average value of ^{232}Th as 40 Bq kg^{-1} in soil, an upward revision by about 60 % as compared to the earlier estimates (NEPA, 1990); which is on par with the current world average value of ^{238}U in soil. With improving knowledge of radioactivity levels in soil, some areas have been identified to have higher ^{232}Th/^{238}U ratio and in extreme situations a ratio up to 15 have been found in some mineral sand areas resulting in higher air exposures of the order of 9.6 μSvh^{-1} (Steinhausler, 1996). ^{220}Rn level is governed by its emanation from soil or building materials containing ^{232}Th, soil characteristics and ambient atmospheric conditions. In terms of radiation protection aspects, major problem for long and short term, measurements of ^{220}Rn arises from the pronounced ^{220}Rn activity concentration gradient which can be found both indoors and outdoors.

Long term profile studies outdoors have shown that its levels vary about 3 orders of magnitude within a range of 3m (Porsendorfer, 1991). Due to spatial variation of ^{220}Rn within a definite measurement volume, the results derived from integrated measurements depends on to a large extend the actual position of the measurement device relative to exhaling surface. ^{220}Rn levels in a dwelling depend mainly on the type of material used for construction. Emanation and ventilation rate, in turn, governs ^{220}Rn levels in dwellings. Exposure to an individual inside a dwelling is mainly due to the external gamma radiation dose received from the primordial nuclides present in the building materials, and the inhalation dose due to ^{222}Rn, ^{220}Rn and their progenies. External gamma exposure from cosmic rays will be less due to shielding effect inside the dwelling. Construction materials and design of the house determine the total exposure. Wide variation in activity content of ^{232}Th in building material is also noticed in different parts (Shukla *et al.,* 1995), are shown in Table 6.

Table 6: ^{232}Th levels in building materials used in India for construction (Menon *et al.,* 1987)

Material	^{232}Th (Bq kg^{-1})
Cement	16 – 377
Brick	21 - 48
Stone	6 - 155
Sand	1 - 5074
Granite	4 - 98
Clay	7 - 1621
Fly ash	7 - 670
Lime stone	1 - 26
Gypsum	7 - 807

Resultant exposure rate varied from 0.04 to 0.79 mSvy^{-1} when the above materials in different proportions are used for construction. ^{220}Rn exhalation rate from soil covers a wide range from 0.27 to 5 Bq m^{-2} s^{-1}, depending on the geology and the emanation characteristics of the ground (Porstendorfer, 1994). In view of this considerable variability, the UNSCEAR has recommended a value of 1 Bq m^{-2} s^{-1}, which appears to unreasonable, since the world average would be close probably to 3 Bq m^{-2} s^{-1}. Tables 7 and 8 respectively gives the reported typical values of ^{232}Th content and ^{220}Rn flux in different types of building materials and typical values of ^{220}Rn exhalation rate in soil and building materials (Steinhausler *et al.,*1994).

Table 7: Typical values of ^{232}Th content and ^{220}Rn flux in different matrices (UNSCEAR, 1982)

Material	^{232}Th content (Bqkg^{-1})	Flux density (Bqm^{-2}s^{-1})
Soil	10-70	0.5 – 5.0
Lime stone	5	0.04
Pumice tone	100	0.5
Black shale	Up to 400	-
Gypsum	10	0.1
Granite	100 - 200	-
Sand stone	5	0.05
Basalt	2 – 15	-
Concrete	25	0.04
Monazite sand	4×10^4 to 3×10^5	0.1

All these, point the need to have a databases on ^{220}Rn levels in indoor air. ^{220}Rn gas is rarely measured, due to the difficulty in measuring an alpha particle emitting gas with a short half-life. It was carried out over a period of two years for ^{220}Rn gas and progeny (^{212}Pb) in floor locations, three indoors and one suburban in outdoor location. An objection to ^{220}Rn gas measurements for dosimetric purpose has been that ^{220}Rn may not be well mixed in the indoor air because of its shorter half-life. Some reported values shows that indoor ^{220}Rn concentrations vary with the distance from the walls and floor (Zhuo, 2001). Indoor measurement results shows that unless the ^{220}Rn detector was located very close to a wall or floor source, the ^{220}Rn was well mixed in room air and provide typical exposure. Large scale surveys of the ^{232}Th concentration in construction materials have been carried out in the past. Reference value of 50 Bq kg^{-1} for ^{232}Th by UNSCEAR (1988) for building materials can be considered as a representative one for the construction materials traditionally used in industrialized countries. Waste materials recycled as building materials; such as bricks made of fly ash or slag, however, have value ranging from 250 to 300 % higher than the UNSCEAR (1988) reference value. No representative value for ^{232}Th data base exists for construction materials used in developing countries. Mean specific activities of ^{232}Th in some building materials (ECRP, 1999; IAEA, 1994) used in several countries are given in Table 9.

Direct measurements of the concentration of all short lived decay products of ^{220}Rn are difficult and limited. Theoretically, the vertical distribution of ^{220}Rn can be predicted fairly well, provided the flux density and eddy-diffusity are known. Even though the subject to research for several decade, the

data base on ^{220}Rn values outdoors is generally not representative one on a global scale, since the data were not of representative one, since the data were not derived from large scale surveys with continuous, long term, time integrating ^{220}Rn measurements (Druilhet, 1992; Reineking, 1992).

Table 8: Typical values of exhalation rate for ^{220}Rn in soil and building materials (UNSCEAR, 1992)

Parameter	Unit	Soil		Building material	
		Range	Typical value	Range	Typical value
Emanation power (ε)	-	0 01 – 0.2	0.05	0.002 – 0.06	0.01
Density (ρ)	10^3 (kgm^{-3})	0.8 – 3.0	2.0	0.1 – 0.25	0.25
Diffusion coefficient(D_b)	(m^2s^{-1})	10^{-8} – 10^{-5}	5×10^{-6}	10^{-8} – 10^{-5}	5×10^{-7}
Diffusion length (R_b)	(m)	0.1 – 0.3	0.02	0.001 – 0.01	0.005
Activity mass conc. (α_b)	($Bqkg^{-1}$)	5 – 120	40	5 – 200	50
Exhalation rate (e)	($Bqm^{-2}s^{-1}$)	10^{-3} – 0.005	1	0.001 – 0.2	0.05

Table 9: Specific activity range of ^{232}Th (Bq kg^{-1}) in some building materials used for construction by different countries (UNSCEAR 2000)

Material	Range (Bq kg^{-1})
Sand	12 – 1008
Cement	44 – 860
Concrete	42 – 918
Tiles	328 – 7541
Bricks	196 – 785
Red bricks	50 – 200
Lime brick	10 – 30
Ceramic	44 – 66
Granite	81 – 87
Marble	11 - 34
Others	2 – 87
Soil	0.05 – 204
Coal fly ash	100 – 300
Gypsum	10 – 100

Concentrations are estimated from the level of equilibrium or disequilibrium between these nuclides and its decay products. An equilibrium factor F_{eq} is defined that permits exposure to be estimated in terms of the potential alpha energy concentration (PAEC) from measurements of ^{220}Rn gas concentration. It has not been practical to assess the lung dose directly from ^{220}Rn gas measurements because the equilibrium factor between the gads and daughters was not well established. Past dose estimates of ^{220}Rn were made from the filtered air measurements of ^{220}Rn decay product ^{212}Pb. However, much measurement results are available from Japan (Tokanami, 1997). It is also not possible to assess the radiation dose from the inhalation of ^{220}Rn decay products by epidemiological studies like ^{222}Rn and therefore it must be estimated using dosimetric modeling. Dosimetric studies (UNSCEAR, 2006) have provided dose conversion factors for assessing the inhalation dose from ^{220}Rn and its progenies both at indoors and outdoors.

In India, data pertaining to ^{222}Rn and ^{220}Rn levels in outdoor, over the ocean and at remote location like Antarctica environment as well as in non uranium mines are available for more than three decades. Recently, in India, the Bhabha Atomic Research Center (BARC) has completed a countrywide monitoring program of ^{220}Rn along with ^{222}Rn in the dwellings using ^{222}Rn-^{220}Rn discriminating Solid State Nuclear Track Detector (SSNTD) based dosimeter. In this presentation, methodology adopted and discusses the results obtained pertaining to ^{220}Rn.

4. Materials and Methods

Several techniques are being used for the measurement of ^{220}Rn in the indoor environment. One has to select the suitable one for the measurement (Monnin, 1989). Conventionally used techniques are either active or passive techniques. Active and passive techniques are used for ^{220}Rn measurements. Some of these techniques are Nuclear Emulsion; Adsorption; Gamma spectrometry; Solid Scintillation; Liquid Scintillation; Beta Monitoring; Solid State Nuclear Track Detector; Ionization Chamber; Surface Barrier Detector; Thermo Luminescent Detect ors; Electrets' dosimeters; Collection Technique; on line continuous monitors; Alpha Guard etc. For the dosimetric point of view, integrated passive technique is preferred since it gives the diurnal, hourly and seasonal variation of ^{220}Rn and its progeny in the indoor environment. A cylindrical

twin cup with small strips of (2.5 × 2.5 cm size) LR 115 Type II 12 µm thick strippable SSNTD films placed on the two compartments and another SSNTD placed outside the chamber as detector. Each compartment of the dosimeter has a length of 4.5 cm and a radius of 3.1 cm as shown in fig.1.

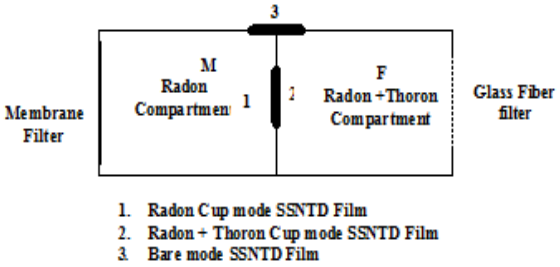

Fig. 1: Schematic diagram of twin cup ^{222}Rn-^{220}Rn dosimeter

Dosimeter is designed, based on the observations in which the efficiency of track production depends on the ratio of overall effective volume to the total volume and that with increase in dimensions of the chamber housing the detector, there is initially a rise in the volume ratio which reaches a maximum and then comes down gradually. Based on these criteria, a cup with the above dimensions has been designed (Bhanthi and Bhagwat, 1996). SSNTD placed in membrane filter compartment measures only ^{222}Rn, which diffuses into the cup from ambient air through a semi-permeable cellulose nitrate membrane sandwiched between glass fiber filter paper, allows > 95 % of the ^{222}Rn gas to diffuse through and due to shorter half life and diffusion properties suppresses ^{220}Rn gas to <1% (Ramachandran *et al.,* 1987).

Mean time for ^{222}Rn to reach a steady state in the cup will be in the range of 4 to 5 hr. In the filter paper and membrane combination mode, which is having a cut off efficiency of 99.8 % for sub µm aerosol particles, the particulates from ^{219}Rn ($T_{1/2}$:3.96 s) and ^{220}Rn ($T_{1/2}$: 55.6 s) will be cut off and will decay while diffusing through the filter paper and membrane combination. ^{222}Rn ($T_{1/2}$:3.82 d) gas, which diffuses through the membrane, will produce the alpha tracks on the detector films placed in this chamber. The SSNTD placed on the other compartment having a glass fiber filter paper, allows both ^{222}Rn and ^{220}Rn gas to diffuse in and hence, the tracks registered on the SSNTD film in this chamber are related to both ^{222}Rn and ^{220}Rn gases. SSNTDs in bare mode (on the outer surface of the dosimeter) register alpha tracks attributable to the airborne concentrations of both the gases and their alpha emitting progeny, namely ^{218}Po, ^{214}Po, ^{216}Po and ^{212}Po. Parameters like the attachment to aerosol, deposition (plate-out), and recoil of ^{222}Rn, ^{220}Rn and their short-lived progenies from aerosols and surfaces, and decay has a major role in the track registration on the bare card detector from ^{222}Rn, ^{220}Rn and their progeny. It is assumed that the SSNTD kept in the bare mode responds to the airborne alpha emitters and not to the alpha activity deposited on it (Ilic and Sutlej, 2000). Studies by Jonsson (1981) have shown that for one hour etching at 60^{o}C, the alpha energy range for the formation of a hole is between 2.2 to 4.1 MeV at normal incidence and the maximum value of the incident angle is about 42^{o}. Upper cut-off energies, hardly changes with the angle of incidence. As a result of this, the alpha emission due to progeny (energy > 5 MeV) deposited on the SSNTDs are not expected to contribute tracks. This supports the assumptions made and is confirmed by experiments by placing a ^{241}Am – ^{239}Pu source (5.48 and 5.15 MeV alpha energies) directly in contact with an SSNTD film and counting the tracks using a spark counter.

Study has shown that the track registration efficiency is negligible due to unsupported activity or un degraded alphas in general and due to this plate out activities in particular. Experiments have shown that the registration efficiency is of the order of 0.001 % (Eappen *et al.,* 1998). Background track density of the SSNTD detectors is important while assessing its performance. Detailed study shows that, sensitivity of the detector exhibits a trend of variation with its age. Variation up to 21 % in the sensitivity of the film from two different batches both processed one-year after the manufacturing has been observed. Twenty five percent increases in the sensitivity factors were observed when these films were recalibrated after a gap of one year. Background track density increases as the age of storage increases from the date of manufacturing. A variation between 2 to 15 track cm^{2} in the background for a storage period of two years was observed. Bare card mode of exposure is also affected by the surface deposition of dust, during the exposure period. Studies carried out to see the effect of dust load on bare card exposure mode, have revealed that the dust collection measured of the order up to 0.3 mg cm^{-2} for a period of 90 days has not tampered the track registration on the detector (Muraleedharan *et al.,* 1993). These dosimeters were deployed into the field on a quarterly cycle of 3 months covering all the seasons in more than 2000 houses of different construction types spread over 50 locations in the country, have been surveyed. After the exposure, the SSNTDs were

retrieved and processed under standard protocols and were scanned under a spark counter to get the total track densities recorded in the bare, filter and membrane compartment. From the total tracks recorded, ^{220}Rn concentration is estimated using the sensitivity factor derived from the controlled experiments (Eappen *et al.,* 2001).

Table 10: ^{220}Rn levels in Dwellings in Literature (Bqm^{-3}) (Porstendorfer, 1994)

Country	Location	No. of data	Mean	Max	Min
Austria	Dwelling	9	19.0	74.0	< 3.3
Brazil	Dwellings	1	19.0	---	---
Germany	Cellars	4	8.9	39.1	2.2
	Lecture room	1	0.7	---	--
	Garage	1	7.6	8.3	4.1
Italy	Dwellings	21	8.5	54.7	---
Sweden	Apartment	---	---	10.0	5.0
	Wooden house	---	---	2.0	1.0
	Basement	---	---	200.0	5.0
	Dwellings	45	31.0	430.0	1.0
Japan	Dwellings	21	8.5	54.7	---
China (HBRA)	Dwellings	---	168.0	---	---
USA	Dwellings	7	10	34	2.0
	Basement	6	13	40	MDL
	Garage	1	10	18	6
	Ground floor	1	12	16	9
India (present work)	Dwellings	1800	12.2	42.2	5.7

Table 11: Reported ^{220}Rn/ ^{222}Rn Progeny Levels in Literature (Porstendorfer, 2004)

Location	^{220}Rn/^{222}Rn Progeny	Comment
Italy (Latium)	1.3	Anomalous (volcanic area), 50 dwellings, poor ventilation
Canada (Elliott Lake)	0.3	Samples at 95 dwellings, source activity ^{238}U/^{232}Th ~ 1
Hungary	0.5	22 dwellings
Norway	0.5	22 dwellings, source activity ^{238}U/^{232}Th ~ 1
FRG (Western part)	0.5	150 measurements spread over an year
FRG (Southwestern)	0.8	95 dwellings
FRG	0.5	27 houses
US	0.6	68 measurements in 20 states
China(Hubei Provinces)	0.4	37 measurements, ^{238}U/^{232}Th ~ 0.6
France (Finistere)	0.3	219 measurements
Hong Kong	0.8	10 indoor sites, a typical tropical coast
Austria	0.7	12 dwellings
UK	0.14	8 dwellings
USA	0.3	53 measurements in 8 south eastern cities on main floor
India(present work)	0.53	1800 houses

5. Results and Discussion

^{220}Rn gas varied from 5.7 to 42.4 Bq m^{-3} with a GM (Geometric Mean) of 12.2 Bq m^{3} (GSD 3.22). Higher ^{220}Rn levels are recorded in some locations where the ^{232}Th content in the surrounding soil is high. Estimated ^{232}Th content in Indian soil using gamma spectrometric techniques varied from 3.5 to 24.7 Bq kg^{-1} with a mean of 18.4 Bq kg^{-1}. It was observed that the ^{232}Th levels in the soil were high in the northern parts of the country (Mishra, 1972).

Table 12: Reported ^{220}Rn levels in some workplaces (Sohrabi, 1997)

Location	Number of Data	Concentration (Bqm^{-3})		
		Min	Max	Mean
Gas mantle factory (UK)	13	1100	11000	-
Mg/Th alloy factories (UK)	-	370	3700	-
Underground U mine (CND)	4	1055	9309	4932
Monazite processing plant (Brazil)	-	-	-	560
Thorium processing plant	2	1800	18000	-
Gas mantle factory, India	8	17	3034	-
^{220}Rn levels in dwellings around some villages of Chavara, Kerala:				
Neendakara	100	8.9	60.7	17.7
Chavara	135	3.9	423.0	27.6
Allappad	120	4.8	76.8	12.4

Table 13: Summary of data on biological effects due to exposure to thorium and its daughter products

Cohort	Exposed persons	Exposure Characteristics	Observed effects
Non-occupational exposure:			
Residents in HBRA:			
Brazil (Paschoa and Pohl-Ruling, 1993)	~ 7000	^{220}Rn levels in air: 0.4 – 19 Bq.m^{-3}	Increased chromosome aberration
China (Wei, 1993)	~ 80,000	^{220}Rn levels indoors : 168 Bq.m^{-3}	Increased chromosome aberration, elevated down's syndrome
India (Sunta, 1993)	~ 70,000	External dose : 7 mGy/y	Increased still birth and infant mortality. Elevated down's syndrome
Occupational exposure:			
Miners of iron ore and Rare earth (Chen *et al.,*1993)	588	Th lung burden: 0.85 Bq	Increased lung cancer incidence. Respiratory diseases
Workers in monazite industry (Lipoztein *et al.,*1992)	300	External dose 14 mSv.y^{-1}	Increased chromosome aberrations
Workers in Thorium processing plant (Polednak *et al.,*1983)	592	Emanating at mouth : 24.5 Bq of ^{224}Ra	Elevated SMR (lung cancer; pancreatic cancer, respiratory diseases)
Medical Exposure:			
German, Japanese and Portuguese (Hofmann, 1988)	~ 53,000	Bronchial life time dose : 357 mGy; Liver dose 2.5 to 3.6 Gy/y	Liver tumors Hepatic tumors

^{232}Th content in materials used for construction in India varied from 124.0 Bq kg^{-1} in sand to 3.1 Bq kg^{-1} in the blue dust (Shukla *et al.,* 1995). Radiation profile map of India have also shows higher levels in the northern parts of the country due to high ^{232}Th content in the rocks since its formation (Shukla *et al.,* 1995), which supports the present observations. Some locations in India are classified as high background radiation areas due to either heavy deposits of monazite or uranium. One such area is located in the southern parts of India (Chavara, Kerala) with high ^{232}Th content in soil. Results of a sample survey carried out in this region shows that in this region the ^{232}Th content in the soil varied from 75 to 9070 Bq kg^{-1} with a mean of 827.0 Bq kg^{-1} (Sankaran *et al.,* 1986), which is 56 times the national average of 18.4 Bq kg^{-1} for the country as a whole excluding the high background radiation regions (Krishnan Nair *et al.,* 1999). Results of a sample survey carried out in nearly 350 dwellings of different types of construction spread over four electoral wards of the high background region (two outside the monazite belt and two near to the monazite belt) around the high background region shows that the indoor ^{220}Rn levels varied from 0.4 to 69.6 Bq m^{-3} with a median of 8.3 Bq m^{-3} in dwellings belonging to the normal background region and from 5.0 to 214.5 Bq m^{-3} with a median of 44.2 Bq m^{-3} in the monazite belt region (Mishra and Sadasivan, 1971).

Using UNSCEAR (2000) dose conversion factors the estimated annual inhalation dose due to ^{220}Rn and its progeny in Indian dwellings around the normal background region excluding high background areas varied from 0.047 to 0.39 mSv y^{-1} with a mean of 0.14 mSv y^{-1} (GSD 1.36). This, when compared with the estimated inhalation dose rates of 1.05 mSv y^{-1} for indoor ^{222}Rn and its progeny in Indian dwellings (Ramachandran *et al.,*1995), total inhalation dose due to ^{220}Rn and its progeny is found to be very small. Inhalation dose rates due to ^{220}Rn and its progeny in dwellings, from the high background region of Chavara, Kerala works out to be 0.41 mSv y^{-1}, which is about 3.2 times higher than that recorded for the country from normal background region. The reported values of annual average potential alpha energy concentrations of radon and thoron progeny in dwellings of Gudalore in South India were 3.54 and 2.65 mWL respectively (Sivakumar, 2010). The reported ^{220}Rn levels in dwellings and work places in available in the literature are given in Table 10 (Muccetelli and Bochicchio, 1998). ^{220}Rn levels measured in Indian dwellings are comparable with those reported for Austria, Brazil and USA. The recorded ^{220}Rn levels in dwellings of China gave a mean concentration of 168 Bq m^{-3} which is 3.5 times the recorded in dwellings from high background regions of India. ^{220}Rn and its progeny can also be significant in underground mines as well as in closed environment. ^{232}Th minerals itself is usually mined from open-air surface deposits. But, they are commonly associated with uranium minerals, so ^{220}Rn exhalation rate from ores of uranium mines is often significant. In addition, due to the possibility of restricted ventilation and proximity to bare soil and rock, any underground mine or enclosures can have significant levels of both ^{222}Rn and ^{220}Rn. If ventilation is not present, underground enclosures can be expected to have ^{220}Rn levels approaching the high values of soil gas. Hence, ventilation, whether natural to manmade, is the key factor, which controls the absolute concentrations of ^{220}Rn and its progeny.

Few data is available for ^{220}Rn gas. Focus is more on ^{220}Rn and its progeny since they are also a significant contributor to total inhalation dose. Biju (1981) reviews on mine models and the ^{220}Rn problem in underground uranium mines this data indicate a median ratio of PAEC (^{220}Rn)/PAEC (^{222}Rn) of 0.65 with a range of about 0.4 to over 1.5. Stranden (1985) and Dixon (1985), have reported measurements on a variety of underground mines and enclosures in Norway and UK. Estimated ratios of PAEC (^{220}Rn)/PAEC (^{222}Rn) were usually in the range of 0.1 to 1.0. Unoccupied mines and natural underground caves will tend have higher values of PAEC (^{220}Rn), but lower value of ratio of PAEC (^{220}Rn)/PAEC (^{222}Rn), due to generally poor ventilation of natural convection. There exists a strong correlation between PAEC (^{220}Rn) and PAEC (^{222}Rn) seems fairly pervasive over a range of housing and locations, although evidences indicate the relation is not a linear one. Study carried out in France by Rannou (1987) has indicated that the phenomenological relation agrees well with the indoor data on ^{220}Rn and ^{222}Rn progeny.

$$\text{PAEC}\,(^{220}\text{Rn}) \propto [\text{PAEC}\,(^{222}\text{Rn})]^{0.4}$$

This relationship is found to be quite consistent. Mean while the average rate was found to be about 0.05 % or higher (Shuai 1999). Houses with high levels of ^{222}Rn progeny will thus have less ^{220}Rn progeny. Although limited measurements of ^{220}Rn in indoor air are available, most investigators have reported both the ^{222}Rn and ^{220}Rn equilibrium equivalent concentrations. This allows some generalizations to be made from derived ratios. Based on the physical characteristics of ^{222}Rn and ^{220}Rn and model entry rates in buildings, ICRP

estimated the expected concentrations in the buildings (ICRP, 1987), this ranged from 10 to 100 Bq m^{-3} for ^{222}Rn and ^{220}Rn both in outdoor air, concrete and brick building materials, and a ventilation rate of 0.7 h^{-1}. In terms of EEC, these indoor concentrations are 2 to 50 Bq m^{-3} for ^{222}Rn and 0.04 to 2 Bq m^{-3} (mean 0.5 Bq m^{-3} for ^{220}Rn). This corresponds to a ^{220}Rn-^{222}Rn EEC ratio of 0.03 (UNSCEAR, 2000).

Table 14: Types of operations involving naturally occurring radioactive materials pertaining ^{232}Th identified as required regulation on the basis of workers dose (IAEA 1994)

Types of Operation	Description	Dominant nuclide	Conc. (Bq g^{-1})	Workers dose (mSv y^{-1})
TiO_2pigment production	Scales during removal from pipes/vessels	^{228}Ra ^{210}Pb	1 to 1600	1 to 6
Thermal Phosphorous production	Fume and precipitator Dust		1000	0.2 to 5
Rare earth extraction from monazite	Monazite Thorium Concentrate Scale Residue	^{232}Th ^{232}Th ^{228}Ra ^{228}Ra	40 to 600 up to 800 1000 23 - 3150	Could approach or exceed dose limit
Production of Thorium compounds	Thorium concentrate Thorium concentrate	^{232}Th ^{232}Th	Up to 800 Up to 2000	Typically 6 to 15 Processing
Manufacture of thorium containing products	Thorium compounds Products	^{232}Th ^{232}Th	Up to 2000 Up to 1000	> 1 to a significant fraction of dose limit
Processing of niobium /tantalum ore	Ore Pyrochlore Concentrate $BaSo_4$ precipitate Slag Preceptor dust	^{232}Th ^{232}Th ^{228}Ra ^{232}Th ^{210}Po	1 to 8 80 200 20 to 120 100 to 500	Could reach a significant fraction of dose limit
Some Underground mines	Ore Scales from Ra rich water	^{232}Th ^{228}Ra	Up to 10 Up to 200	< 1 to a significant fraction of dose limit
Oil and gas production	Scales during removal from pipes/vessels	^{228}Ra	0.1 to 15000	< 1 to a significant fraction of dose limit
Fused Zirconia production	Do	^{210}Po	Up to 600	0.25 to 3

Table 11 gives the rounded values of means or medians of the reported ratio of potential alpha energy concentration of ^{220}Rn to that of ^{222}Rn progeny for various locations. In India, lamps using thoriated gas mantles are still used for indoor and outdoor lighting in homes and hawkers in rural as well as urban areas. Presently there are about 75 manufacturing units handling on an average about 200 metric tons of thorium nitrate per annum in the manufacturing of gas mantles in the country. On an average 200 million mantles are made per year, from which 25 % are exported. Considering the large quantities being handled contribution to the inhalation dose of the workers from the ^{220}Rn gas emanated and build up of the progeny in ambient air may also be significant. As per the regulatory body specification, the quantity of thorium allowed in a gas mantle depends on its luminous intensity.

Table 15: Naturally occurring radioactive material that is considered for regulation pertaining ^{232}Th on the basis of activity concentration (IAEA, 1994)

Material Category	Material	Predominant Nuclide	Typical Activity (Bq g^{-1})
Raw material	Monazite sand	^{232}Th	40 – 600
	Metal ore	^{232}Th	up to 10
	Bauxite	^{232}Th	0.035 – 1.4
Products	Gas mantle	232Th	500 – 1000
	Thoriated glass	^{232}Th	200 – 1000
	Thorium containing optical polishing powders	^{232}Th	150
	Thoriated welding electrodes	^{232}Th	30 – 150
	Thorium alloys	^{232}Th	46 – 70
	Zircon refractories	^{238}U	0.14 - 2
Slag	Niobium extraction	^{232}Th	20 to 120
	Tin smelting	^{232}Th	0.07 – 15
Scales, Sludges, sediments	$BaSO_4$ precipitate	^{232}Th	200

Permitted quantity of thorium in a mantle of up to 400 cd rating is 600 mg and for greater than 400 cd, it is 800 mg (Mantle industry continuous to specify the rating in candle power which is equivalent to candela – cd in SI units) (Sadagopan *et al.,* 1997). Besides, in the high background areas of Chavara, Kerala, inhalation exposure due to ^{220}Rn and its progeny is also high. Table 12 gives the reported ^{220}Rn levels in some industries across the world (Paschoa and Pohl-Ruling, 1993). From this Table it can be seen that other than dwelling environment, other workplaces like gas mantle and monazite processing industry and thorium processing plant also have higher ^{220}Rn exposures. Table 13 summarizes major findings on the data on the biological effects among humans due to exposure to ^{232}Th and decay products. Here the main exposure pathways are non-occupational exposure to ^{220}Rn and decay products; occupational exposure to natural thorium and medical exposure to thorium oxide. Here the major studies were confined to three fields: (i) Non-occupational exposure to ^{220}Rn and decay products. This exposure is continuous, low-level exposure associated with increased chromosome aberration, changes in the fertility history parameters and Down's syndrome at atmospheric levels > or equal to 168 Bq m^{-3} at an average external dose rate of about 7 mGy y^{-1}; (ii) Occupational exposure to natural thorium confined to long term, elevated exposure of industrial workers and miners resulted in the incidence of increased chromosome aberration, pancreatic cancer and respiratory diseases (average Th lung burden : 0.85 Bq; average lower dose 9.4 Gy; ^{224}Ra emanation from the mouth greater than or equal to 24.5 Bq); and (iii) medical exposure to thorium oxide confining to injection of thorotrast resulted in lifetime excess cancer risk (bone, liver) and leukemia, ranging from 55 to 330 per 10^4 persons per Gy (BEIR-IV, 1988).

Radionuclide of natural origin is ubiquitous in the environment at variable, but generally low, activity concentrations. Regulation of human activities involving material containing these radionuclide at activity concentrations that would invoke widespread regulatory consideration, in circumstances where it is unlikely to achieve any improvement in protection, would be an optimum use of regulatory resources. So, values of activity concentrations in materials 1 Bq g^{-1} for ^{238}U and ^{232}Th and 10 Bq g^{-1} for ^{40}K are specified in the standards as being values below which it is usually unnecessary to regulate, irrespective of the quantity of material or whether it is in its natural state or has been subject to some form of processing (Zhuo *et al.,* 2002). Table 14 gives the types of operations involving naturally occurring radioactive materials pertaining to ^{232}Th, identified as required regulation on the basis of worker dose. Table 15 gives the Naturally Occurring Radioactive Material pertaining to ^{232}Th on the basis of the activity concentration reported in literature (IAEA, 1994).

6. Conclusions

Based on the literature and the data generated in India the following conclusions are being projected (a) ^{220}Rn levels in Indian dwellings varied from 5.7 to 42.2 Bq m^{-3} with a GM of 12.2 Bq m^{-3} (b) Higher ^{220}Rn levels are recorded in dwellings around locations where the ^{232}Th content in soil also high (c) Estimated national average value of ^{220}Rn levels

for India are comparable with those reported for Austria, Brazil and USA (d) Inhalation dose rate due to ^{220}Rn and its progeny varied from 0.047 to 0.39 mSv y^{-1} with a GM of 0.14 mSv y^{-1} (e) Inhalation dose rate due to ^{220}Rn and its progeny in dwellings from high background regions were found to be nearly 3.2 times higher than those recorded in dwellings around normal background regions in India (f) Estimated ratio of ^{220}Rn/^{222}Rn progeny levels in Indian dwellings works out to be 0.93, which lie in the range 0.3 to 1.0 reported from different countries all over the world. Preliminary indoor surveys carried out in some western countries are indicative of a non negligible ^{220}Rn/^{222}Rn exposure component for some members of the general public. This is also the case with some occupational exposure received at some work places, particularly in monazite processing industry. But the presently available data on ^{220}Rn/^{222}Rn daughter levels, aerosol characteristics, and their behavior outdoors and indoors cannot be considered as representative.

There is also added ambiguity in the current understanding of potential health determinants due to the lack of any established dose effect relationship and contradictory evidence of biological effects induced by ^{220}Rn/^{222}Rn daughters. Efforts should be warranted to address the ^{220}Rn/ ^{222}Rn daughter issue on an international scale. Emphasis should be on such areas which will assist in improving the current dose assessment of population groups estimated to receive partly significantly elevated ^{220}Rn/ ^{222}Rn daughter exposures. In summary, our scientific knowledge is such that, on an average, worldwide ^{220}Rn is expected to be less of a problem than ^{222}Rn. In view of limited resources, research work should be focused first of all on identification of problem situation and improve the estimate of overall contribution from ^{220}Rn. This would represent a less expensive goal than the broad scaled search and rescue operations like, the one, which occurs with ^{222}Rn. Possible role of exposures to ^{220}Rn and its daughter products is of increasing interest, and a number of research workers have reported that ^{220}Rn can be detected as a significant component of the total ^{222}Rn + ^{220}Rn; ^{220}Rn can thus be a source of error in residential ^{222}Rn studies which do not distinguish the two contributions to exposure (Zhuo *et al.,* 2002). Further studies are needed to consider the contribution of both ^{222}Rn and ^{220}Rn.

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