^{220}Rn IN INDOOR ENVIRONMENT AND WORK PLACES: 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 given. Results are compared with the values reported in literature for dwellings as well as in high background radiation areas.

Keywords

India, indoor, inhalation dose, SSNTD, Thoron.

Introduction

Topics 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 to chronic low levels. 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 becomes necessary , when calculating dose, to supplement the external component with an inhalation component. This component is not adequately estimated for in any country so far on a national level. The ^{220}Rn problem will also be a problem in industries which use 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 amounts of thorium nitrate are being handled by these industries, the 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 increasingly felt that it may be necessary to have data on the ^{220}Rn in the environment for obtaining a complete picture of the inhalation dose. In this paper, the current status of ^{220}Rn levels in the indoor environment

in workplaces as well as in other industries where large amount of ^{232}Th is being handled is summarized. Methods of measurement and reported levels in literature are also summarized.

Succinct 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 has been generated on the levels of ^{222}Rn in the environment (UNSCEAR, 2000). In contrast, data on ^{220}Rn is scarce 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 which observed high ^{220}Rn in the living environments in various countries and it 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 (Porstendorfer, 1994; Steinhausler et al., 1994).

^{220}Rn was 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 considerations.

An 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 ^{the} formation of thunderstorms. ^{220}Rn and its progeny have been used as a 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, which play 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 since wastes and tailings from ^{232}Th bearing ores processed for metals other than ^{232}Th can potentially release significant ^{220}Rn.

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 et al., 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 Bqkg^{-1} (UNSCEAR, 2000). With organic matter, there is some tendency for 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, pegmatities 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 rise to a higher inhalation dose rate to the public. The decay scheme of the ^{232}Th series is given in Table 1.

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	-	-	-

A number of locations with higher content of ^{232}Th have 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, $<10^{-4}$ Bqkg^{-1} (Wedepohl, 1978). ^{232}Th content in the soil around these high background areas varied from 0.5 to 1000 Bqkg^{-1} (UNSCEAR, 2006).

The basic physical properties of ^{220}Rn are given in Table 2. The immediate parent of ^{232}Th is ^{224}Ra.

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

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

This isotope, ^{224}Ra, is not always in equilibrium with ^{232}Th, particularly in ground water. However, in broad terms its concentration in soils and rocks will correlate well with ^{232}Th. Upon ^{224}Ra decay, the ^{220}Rn atom will experience recoil. If decay takes place within a mineral, the recoil range is of the order of 30nm. 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.

Table 3: Common values of ^{232}Th content and ^{220}Rn flux density (Stenhausler, 1996)

Material	^{232}Th ($Bqkg^{-1}$)	Flux density ($Bqm^{-2}s^{-1}$)
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	-

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, 2005) and is given in Table 4.

Table 4: Estimated ^{232}Th reserves (tones of Thorium metal) (IAEA, 2005)

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 (MTs) respectively. ^{232}Th deposits in India has been recently reported to be in the range of 0.65 MTs. Large known reserves 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 mainly 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 Table 5.

Table 5: Typical radioactivity content of beach sands and monazite of Indian continent (IAEA, 2005)

	Activity concentration (Bqkg^{-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 the decay chain up to ^{214} Po contribute 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 positively charged state. If unattached to aerosols, these, usually existing as molecular clusters, have a diffusion coefficient in air about 0.05 cm^2s^{-1}, with the exact value depending on the properties of the air 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. ^{220}Rn can migrate to the earth's atmosphere and can be inhaled along with its progeny. ^{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 $Bqkg^{-1}$ 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, measurable amounts of ^{220}Rn arise 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 with in 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 to a large extent on 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, govern ^{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 the shielding effect inside the dwelling. Construction materials and design of the house determine the total exposure. Wide variations in the activity of ^{232}Th in building material is also noticed in (Shukla, 1995) as shown in Table 6.

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

Material	^{232}Th ($Bqkg^{-1}$)
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 rates 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 $Bqm^{-2}s^{-1}$ 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 $Bqm^{-2}s^{-1}$, which appears to unreasonable, since the world average would be close probably to 3 $Bqm^{-2}s^{-1}$. 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 (Stenhausler, 1994)

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(black)	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

Table 8: Typical values of exhalation rate for ^{220}Rn in soil and building materials (Stenhausler, 1994)

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 con. (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

These graphs seem to point out the need to have a database 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. Another 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. The reference value of 50 $Bqkg^{-1}$ for ^{232}Th by UNSCEAR (1988) for building materials can be considered as a representative 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 values ranging from 250 to 300% higher than the UNSCEAR (1988) reference value. No representative value for ^{232}Th data 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.

Table 9: Specific activity range of ^{232}Th in some building materials used for construction by different countries (IAEA, 1994)

Material	Range (Bqkg^{-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

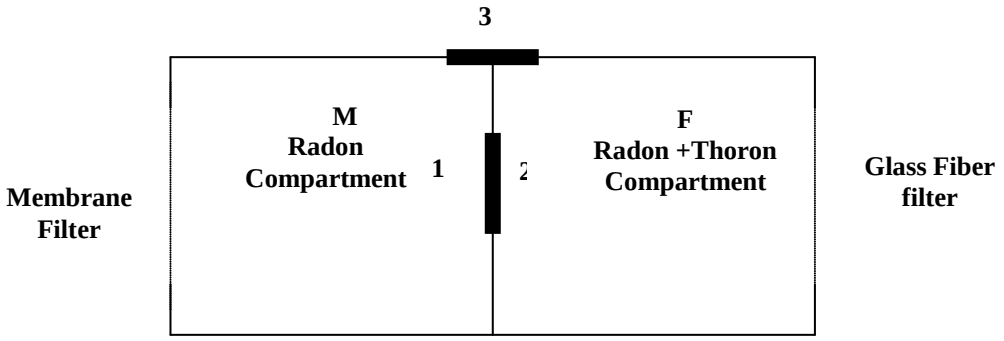
Direct measurements of the concentration of all short lived decay products of ^{220}Rn are difficult and limited. Theoretically, the vertical distribution of ^{220}Rn can be predicted fairly well, provided the flux density and eddy-diffusivity are known. Even though the subject of research for several decades, the data base on ^{220}Rn values outdoors is generally not a representative one on a global scale, since the data were not derived from large scale surveys with continuous, long term, time integrating ^{220}Rn measurements (Druilhet, 1992; Reineking, 1992).

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 ^{220}Rn and its decay products was not well established. Past dose estimates of ^{220}Rn were made from the filtered air measurements of the ^{220}Rn decay product ^{212}Pb. Also, some 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 there fore 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 outdoor, over the ocean and at remote location like Antarctica environment as well as in non-uranium mines have been 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, the methodology adopted and the results obtained pertaining to ^{220}Rn are discussed.

Measurement Methodology

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. 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 Detectors, Electret dosimeters, Collection Technique and continuous monitors (Alpha Guard etc.) For the dosimetric point of view, the 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 is used 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 a detector. Each compartment of the dosimeter has a length of 4.5 cm and a radius of 3.1 cm as shown in figure 1.



1. **Radon Cup mode SSNTD Film**
2. **Radon + Thoron Cup mode SSNTD Film**
3. **Bare mode SSNTD Film**

Fig. 1: Schematic of twin cup ^{222}Rn-^{220}Rn dosimeter developed by Bhabha Atomic Research Center, Mumbai, India

The dosimeter is designed, based on the observation that the efficiency of track production depends on the ratio of over all effective volume to the total volume and that with an 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). A SSNTD placed in a 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, allowing >95% of the ^{222}Rn gas to diffuse through and, due to the shorter half life and diffusion properties, suppresses ^{220}Rn gas to <1% (Ramachandran et al., 1987).

The 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 has 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 Sutej, 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.

Experiments have shown that the registration efficiency is of the order of 0.001% (Eappen and Mayya 2004). 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 on the order of up to 0.3 mgcm^{-2} for a period of 90 days has not affected the track registration on the detector (Muraleedharan et al., 1993).

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. 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).

Results

^{220}Rn gas concentration varied from 5.7 to 42.4 Bqm^{-3} with a GM (Geometric Mean) of 12.2 Bqm^{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 Bqkg $^{-1}$ with a mean of 18.4 Bqkg^{-1}. It was observed that the ^{232}Th levels in the soil were high in the northern parts of the country (Mishra, 1972). ^{232}Th content in materials used for construction in India varied from 124.0

$Bqkg^{-1}$ in sand to 3.1 $Bqkg^{-1}$ in the blue dust (Shukla et al., 1995). A radiation profile map of India also shows higher levels in the northern parts of the country due to high ^{232}Th content in the rocks (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 the ^{232}Th content in the soil varied from 75 to 9070 $Bqkg^{-1}$ with a mean of 827.0 $Bqkg^{-1}$ (Sankaran et al., 1986), which is 56 times the national average of 18.4 $Bqkg^{-1}$ 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 in the high background region (two outside the monazite belt and two near to the monazite belt) shows that the indoor ^{220}Rn levels varied from 0.4 to 69.6 Bqm^{-3} , with a median of 8.3 Bqm^{-3} in dwellings belonging to the normal background region and from 5.0 to 214.5 Bqm^{-3} with a median of 44.2 Bqm^{-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 varied from 0.047 to 0.39 $mSvy^{-1}$ with a mean of 0.14 $mSvy^{-1}$ (GSD 1.36). This, when compared with the estimated inhalation dose rates of 1.05 $mSvy^{-1}$ for indoor ^{222}Rn and its progeny in Indian dwellings (Ramachandran et al., 1995), implies a total inhalation dose due to ^{220}Rn and its progeny to be very small. Inhalation dose rates due to ^{220}Rn and its progeny in dwellings, from the high background region of Chavara, Kerala work out to be 0.41 $mSvy^{-1}$, which is about 3.2 times higher than that recorded for the country from normal background region. The reported ^{220}Rn levels (Bqm^{-3}) in dwellings and work places 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 Bqm^{-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 a closed environment. The ^{232}Th mineral 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.

Table 10: ^{220}Rn levels in Dwellings in Literature (Muccetelli et al., 1998)

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

Hence, ventilation, whether natural or man made, is the key factor which controls the absolute concentrations of ^{220}Rn and its progeny. Few data are available for ^{220}Rn gas alone. The focus is more on ^{220}Rn progeny since they may be a significant contributor to total inhalation dose. Biju (1981) reviews some mine models and the ^{220}Rn problem in underground uranium mines. These 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 to have higher values of PAEC (^{220}Rn), but lower values of the ratio of PAEC (^{220}Rn) / PAEC (^{222}Rn), due to generally poor ventilation from 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 evidence indicates the relation is not a linear one. A study carried out in France by Rannou (1987) has indicated that the phenomenological relation PAEC (^{220}Rn) $\propto$ [PAEC (^{222}Rn)] $^{0.4}$ agrees well with the indoor data on ^{220}Rn and ^{222}Rn progeny. This relationship is found to be quite consistent. 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 Bqm^{-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 Bqm^{-3} for ^{222}Rn and 0.04 to 2 Bqm^{-3} (mean 0.5 Bqm^{-3} for ^{220}Rn). This corresponds to a ^{220}Rn-^{222}Rn EEC ratio of 0.03 (UNSCEAR, 2000). 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 the build-up of the progeny in ambient air may also be significant.

Table 11: Reported $^{220}Rn/^{222}Rn$ Progeny Levels in Literature (Rannou, 1987)

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

As per the regulatory body specification, the quantity of thorium allowed in a gas mantle depends on its luminous intensity. The 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 (The mantle industry continuous to specify the rating in candle power which is equivalent to candela – cd in SI units) (Sadagopan et al., 1997).

In addition, 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).

Table 12: Reported ^{220}Rn levels in some workplaces (Paschoa, 1993)

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

From this Table it can be seen that other than the dwelling environment, other workplaces like gas mantle and monazite processing industry and thorium processing plants 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. These studies were confined to three fields: (i) Non-occupational exposure to ^{220}Rn and decay products, (ii) Occupational exposure to natural thorium confined to long term, elevated exposure of industrial workers and (iii) medical exposure to thorium oxide confined to injections of thorotrast.

Table 13: Summary of data on biological effects due to exposure to thorium and its daughter products (IAEA, 2005)

Cohort	No. of exposed persons	Exposure Characteristics	Observed effects
Residents in HBRA in:			
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

Radionuclides of natural origin are ubiquitous in the environment at variable, but generally low, activity concentrations. Regulation of human activities involving material containing these radionuclides 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 Bqg^{-1} for ^{238}U and ^{232}Th and 10 Bqg^{-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., 2001). 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 (Zhuo et al., 2001).

Table 14: Types of operations involving naturally occurring radioactive materials pertaining ^{232}Th identified as required regulation on the basis of workers dose (IAEA, 2005)

Types of Operation	Description	Dominant nuclide	Conc. (Bqg^{-1})	Workers dose ($mSvy^{-1}$)
TiO_2 pigment 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	^{232}Th	40 to 600	Could approach or exceed dose limit
	Thorium Concentrate	^{232}Th	up to 800	
	Scale	^{228}Ra	1000	
	Residue	^{228}Ra	23 - 3150	
Production of Thorium compounds	Thorium concentrate	^{232}Th	Up to 800	Typically
	Thorium concentrate	^{232}Th	Up to 2000	6 to 15 Processing
Manufacture of thorium containing products	Thorium compounds	^{232}Th	Up to 2000	> 1 to a significant fraction of dose limit
	Products	^{232}Th	Up to 1000	
Processing of niobium /tantalum ore	Ore	^{232}Th	1 to 8	Could reach a significant fraction of dose limit
	Pyrochlore Concentrate	^{232}Th	80	
	$BaSo_4$ precipitate			
	Slag	^{228}Ra	200	
	Preceptor dust	^{232}Th	20 to 120	
		^{210}Po	100 to 500	
Some Underground mines	Ore	^{232}Th	Up to 10	< 1 to a significant fraction of dose limit
	Scales from Ra rich water	^{228}Ra	Up to 200	
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 15: Naturally occurring radioactive material that is considered for regulation pertaining to ^{232}Th on the basis of activity concentration (Zhuo, 2001)

Material Category	Material	Predominant Nuclide	Typical Activity (Bqg^{-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

Findings

Based on the literature and the data generated in India, the following has been found:

a. ^{220}Rn levels in Indian dwellings varied from 5.7 to 42.2 Bqm^{-3} with a GM of 12.2 Bqm^{-3} .
b. Higher ^{220}Rn levels are recorded in dwellings around locations where the ^{232}Th content in soil also high.
c. The estimated national average value of ^{220}Rn levels for India are comparable with those reported for Austria, Brazil and USA.
d. The inhalation dose rate due to ^{220}Rn and its progeny varied from 0.047 to 0.39 $mSvy^{-1}$ with a GM of 0.14 $mSvy^{-1}$.
e. The 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. The estimated ratio of ^{220}Rn/^{222}Rn progeny levels in Indian dwellings works out to be 0.93, which lies within the range of 0.3 to 1.0 reported from different countries all over the world.

Conclusions

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 progeny issue on an international scale. Emphasis should be on such areas which will assist in improving the current dose assessment of population groups expected to receive significantly elevated ^{220}Rn/ ^{222}Rn daughter exposures. In view of limited resources, research work should be focused, first of all, on identification of specific situations expected to be problematic and second of all, to 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.

Exposures to ^{220}Rn and its decay 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 in the environment; ^{220}Rn can thus be a source of error in residential ^{222}Rn measurement studies which do not distinguish the two contributions to exposure (Zhuo et al., 2002).

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