

• *Review article*

Environmental thoron (^{220}Rn) : A review

T.V. Ramachandran

19-A/201, Verain Regency Estate, Kalyan _ Shill Road, Dombivli (E) 421 202, Maharashtra, India

Ever since studies on uranium miners 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, there was a great upsurge of interest in the measurement of ^{222}Rn in the environment and considerable data is generated on the levels of ^{222}Rn in the environment across the worlds and is periodically reported by UNSCEAR. In contrast to this, data pertaining to ^{220}Rn in indoors and workplace environment is scarce 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. Many locations have higher levels of natural background radiation due to elevated levels of primordial radio-nuclides in the soil and their decay products like radon (^{222}Rn), and thoron (^{220}Rn) in the environment. It is estimated 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. 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 threated 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. In this article 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. **Iran. J. Radiat. Res., 2010; 8 (3): 129-147**

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INTRODUCTION

Ever since studies on uranium miners 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, 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 ⁽¹⁾. In contrast, data on ^{220}Rn is scarce due to the general perception that its level is negligible due to its shorter $T_{1/2}$ (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 it is now 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 ⁽¹⁻⁴⁾.

The two main reasons for this unsatisfactory situations are: 1) convenient, low cost time integrated measuring devices for ^{220}Rn , suitable for large-scale screening surveys, are unavailable; and 2) based on the only few, frequently non-representative $^{220}\text{Rn}/^{222}\text{Rn}$ -d surveys carried out in the past, it was concluded that further efforts were not warranted, since these data showed apparently – by comparison with ^{222}Rn – only a minor contribution to the overall dose from natural radiation environment. As it is known now, the conclusions about the lack of importance of $^{220}\text{Rn}/^{222}\text{Rn}$ – d may have been premature, since the exposure scenarios have been identified, where ^{220}Rn and ^{220}Rn -d exposure may represent a significant fraction of the total natural radiation exposure to the general

***Corresponding author:**

Dr. T.V. Ramachandran,
19-A/201, Verain Regency Estate, Kalyan _ Shill
Road, Dombivli (E) 421 202, Maharashtra, India
E-mail: torradon@gmail.com

population indoors due to the use of undesirable construction materials. Furthermore, scenarios have been identified for occupational exposure as a result of mining, milling and waste management of heavy mineral sands ⁽⁵⁾. In some cases, the effective dose is also comparable to that of ²²²Rn-d ⁽⁶⁾.

Thoron (²²⁰Rn) was discovered in 1899 by R.B.Owens 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 ²²⁰Rn research occurred in the atmospheric sciences when it was realized that ²²⁰Rn and its progeny are a major source of atmospheric ions near the earth's surface. These ions are important to a wide range of atmospheric processes which are necessary for radon and formation of thunderstorms. ²²⁰Rn and its progeny have been used as tracer in studies of atmospheric transport processes, such as eddy diffusion. Much of these early atmospheric research was by Israel and others ^(7, 8) and the field has continued to be very active ⁽⁹⁾. There are few industrial situations where ²²⁰Rn can be found to be more in isolation from ²²²Rn. Most of these are connected with industrial applications of thorium. Thorium is a component in certain alloys, like magnesium, and plays a small role in nuclear fuel cycle industry. Health problems associated with these applications have not been striking. Mining of thorium ore is done in well ventilated open pits. Wastes and tailings from thorium bearing ores processed for metals other than thorium can potentially release significant ²²⁰Rn.

Review of physical properties

Thorium (²³²Th) is the ultimate progenitor of ²²⁰Rn, its distribution in the earth's crust is important for controlling the production of ²²⁰Rn. Trace amount of ²³²Th permeate almost all soils and rocks, in part

due to the influence of ground water from which thorium can precipitate over geological time scales. ²³²Th usually exist in plus four valence state. It is not highly soluble itself, but forms complex ions which are more soluble ⁽¹⁰⁾. ²³²Th 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 ²³²Th is not as soluble as ²³⁸U, there is some similarity in their geochemistry, and soils enhanced in ²³⁸U are often enhanced in ²³²Th. In magmas and hard rocks, there is likely to be an even stronger correlation between ²³²Th and ²³⁸U deposits since here the respective geochemical processes have a greater similarity yet.

Average concentration of thorium in soil is estimated to be 25 Bq.kg⁻¹ ⁽¹⁾, with organic matter, there is some tendency with unusually high ²³²Th content including monazite, thorite, zircon, sphene, and allanite. Rocks composed of granite or black shale are likely to have high thorium content. Monazite and zircon sands have an especially high concentration of thorium. In contrast, basalt, lime stone, and sand stone typically have a below average concentration. Thorium 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 thorium is present as nearly 100 % thorium isotope. In general, thorium occurs in association with uranium and rare earth element (REE) in diverse rock types; as veins of thorite, thorianite, uranothorite and as monazite in granites, syenites, pegmatites and other acidic intrusions. Monazite also present in quartz-pebble conglomerates sand stones and in fluvial and beach placers. Thorium is also found as an associate element with REE bearing bastnaesite in carbonates.

Present knowledge of thorium resources is limited and incomplete due to the relatively low-key exploration efforts arising out of insignificant demand. Apart

from its main use in nuclear energy, as fertile material, thorium 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 H²), throat tungsten welding rods and in magnesium-based alloys.

All these applications give raise higher inhalation dose rate to the public. Number of locations with higher content of ²³²Th 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 ²³²Th. In contrast, the ²³²Th content of the oceans far from freshwater discharge is quite low, less than 10⁻⁴ Bq.kg⁻¹ (30). ²³²Th content in soil around this high background areas varied from 0.5 to 1000 Bq kg⁻¹(23). Table 1 gives decay scheme of the thorium series. Table 2 gives the properties of ²²⁰Rn.

Table 3 gives typical values of thorium content and thoron flux density in different materials. Immediate parent of ²³²Th is

²²⁴Ra.. This isotope is not always in equilibrium with ²³²Th, particularly in ground water, in broad terms its concentration in soils and rocks will correlate well with ²³²Th. Upon decay ²²⁴Ra, the ²²⁰Rn atom will experience recoil. If decay takes place within a mineral, the recoil range is of the order of 30 nm. So ²²⁰Rn atoms might be expected to remain trapped in the grains for the short time they exist before decay. Typical value for ²²⁰Rn concentration in the pore air of deep soil is estimated to about 20000 Bq.m⁻³ (corresponding to a soil with about 25 Bq.kg⁻¹ ²³²Th, porosity 50 %, density 1.5 g.cm⁻³ and an emanation coefficient of 0.3). Known world reserve of thorium in reasonably assured reserves (RAR) and

Table 2. Properties of ²²⁰Rn.

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

Table 1. Principal members of the thorium series.

Nuclide	Half-life	Major radiations 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			

estimated additional reserve (EAR) categories are in the range of 2.23 MT and 2.13 MT respectively ⁽¹¹⁾ and are given in table 4.

In the RAR category, the deposits in Brazil, Turkey and India are in the range of 0.60, 0.38 and 0.32 MT respectively. Thorium deposits in India has been recently reported to be in the range of 0.65 MTs. Large known reserve of thorium are contained in the beach sand and inland placer deposits of monazite, a mixture of phosphate minerals with chemical formula (RE/TH/U) PO₄. Monazite is a primary source of light REE and thorium and a secondary source of phosphate and uranium. Hazards from thorium 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 thorium bearing dust and short-lived decay products of thoron gas (²²⁰Rn). Table 5 gives activity content of beach sands and monazite of Indian continent.

Radon isotopes are inert gases which form chemical compounds only with difficulty ⁽¹²⁾. ²²⁰Rn progeny in decay chain up to ²⁰⁸Tl 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 cm² s⁻¹, with the exact value depending on the properties of the air like moisture content and the presence of trace gases ⁽⁴⁾.

Existence of high ²²⁰Rn levels were already investigated thoroughly where ever surveys were carried out with the ²²²Rn - ²²⁰Rn discriminative measurements, behavior of the ²²⁰Rn and its progenies and their effects on human health have not been clearly elucidated yet. ²²⁰Rn can migrate to earth's atmosphere exhibit with indoors and outdoors, can be inhaled mainly its progeny, through inhalation. ²²⁰Rn is a natural production of ²³²Th series in the earth's crust like soil, rocks and also in building materials ⁽¹³⁾. Estimates show a range of values for ²³²Th levels in the ground surface. UNSCEAR ⁽¹⁾ has estimated a world average value of ²³²Th as 40 Bq kg⁻¹ in soil, an

Table 3. Common values of thorium content and thoron flux density.

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 ²³²Th reserves (tones of Th metal).

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

upward revision by about %60 as compared to the earlier estimates ^(14, 15); 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 Th / 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 mSv/h ⁽¹⁶⁾.

^{220}Rn level is governed by its emanation from the soil or building materials containing ^{232}Th , soil characteristics and ambient atmospheric conditions. In terms of radiation protection aspects, a major problem for long term 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 the ^{220}Rn levels vary about 3 orders of magnitude with in a range of 3 m ⁽¹⁷⁾. Due to the significant spatial variation of ^{220}Rn within a definite measurement volume, the results derived from integrated measurements mainly 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. Radiation 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 radioactivity content due to ^{232}Th in building material is also noticed (table 6) in different parts ⁽¹⁸⁾.

Resultant exposure rate varied from 0.04 to 0.79 mSv/y 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 ⁽¹⁹⁾. 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. Table 7 and 8 respectively gives the reported typical values of thorium content and thoron flux in different types of building materials and typical values of ^{220}Rn exhalation rate in soil and building materials ⁽³⁾.

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 both ^{220}Rn gas and progeny (^{212}Pb) in floor locations, three indoors and one suburban n outdoor location. An objection to ^{220}Rn gas measurements for dosimetric purpose has been that

Table 5. Typical radioactivity content of beach sands and monazite of Indian continent.

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

Table 6. ^{232}Th levels in building materials used in India for construction.

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

Table 7. Typical values of thorium content and thoron flux in different matrices.

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

Table 8. Typical values of exhalation rate for thoron in soil and building materials.

Parameter	Unit	Soil		Building material	
		Range	Typical value	range	Typical value
Emanation power (e)	-	0.01 – 0.2	0.05	0.002 – 0.06	0.01
Density (r)	10 ³ (kg/m ³)	0.8 – 3.0	2.0	0.1 – 0.25	0.25
Diffusion coeff.(D _b)	(m ² /s)	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. (a _b)	(Bq/kg)	5 – 120	40	5 - 200	50
Exhalation rate (e)	(Bq/m ² /s)	10 ⁻³ – 0.005	1	0.001 – 0.2	0.05

²²⁰Rn may not be well mixed in the indoor air because of its shorter half-life. Reported values show that indoor ²²⁰Rn levels vary with the distance from the walls and floor (20). Results show that unless the ²²⁰Rn detector was located very close to a wall or floor source, the ²²⁰Rn was well mixed in room air and provide typical exposure. Several large scale surveys of the ²³²Th concentration in construction materials have been carried out in the past. Reference value of 50 Bq kg⁻¹ for ²³²Th by UNSCEAR (21) 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 average value ranging from 250 to 300 % higher than the reference value. No representative value for ²³²Th data base exists for construction materials used in developing countries. Mean specific activities of ²³²Th in some

building materials used in several countries is given in table 9 (22-25). Direct measurements of the concentration of all short lived decay products of ²²⁰Rn are difficult and limited. Theoretically, the vertical distribution of ²²⁰Rn can be predicted fairly well, provided the flux density and eddy-diffusivity are known. Even though the subject to research for several decade, the data base on ²²⁰Rn values outdoors is generally not representative one on a global scale, since the data were not of representative on a global scale, since the data were not derived from large scale surveys with continuous, long term, time integrating ²²⁰Rn measurements (26-29).

Levels are estimated from the level of equilibrium or disequilibrium between these nuclides and its decay products. Equilibrium factor F_{eq} is defined that permits exposure to be estimated in terms of the potential alpha energy concentration (PAEC) from measurements of ²²⁰Rn gas

Table 11. Methods of measurements of thoron progeny in WL units.

Sr. No	Method	Measurement		MDL	Remarks
1	One count filter paper sampling	active	30 to 60 min	0.1 mWL	Error of measurement (10 %)
2	Least square analysis of decay of filter paper sample	active	A few min to few hr	1.0 mWL	Error of measurement (1 – 2 %)
3	SSNTD(BARE)	passive	1 to 3 month	0.6 mWL	RSD 20 %

Table 12. Methods of measurement of thoron progeny in presence of a mixture of radon and thoron.

Sr. No	Method	Measurement		MDL (Bq m ⁻³ or mWL)	Remarks
1	Double filter	Active	30 to 60 min	2.2 for thoron	-
2	Filter paper sample of progeny	active	Few min to few hr	< 1 Bqm ⁻³ for thoron and 1 mWL for thoron progeny	-
3	SSNTD	passive	1 to 3 month	1.2 Bq m ⁻³ for thoron and 0.63 mWL for thoron progeny	-

Since the dosimetric point of view, integrated passive technique is preferred since it gives the diurnal and hourly and seasonal variation of ²²⁰Rn and its progeny in the indoor environment. We have used a twin cup cylindrical one with small strips of (2.5 × 2.5 cm size) Kodak LR 115 Type II 12 mm 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. Dosimeter is designed, based on the observations in which the efficiency of track production depends on the ratio of over all 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 (36, 37).

SSNTD placed in membrane filter compartment measures only ²²²Rn, which diffuses into the cup from ambient air through a semi-permeable cellulose nitrate membrane sandwiched between glass fiber filter paper, allows more than 95 % of the ²²²Rn gas to diffuse through and due to shorter half life and diffusion properties suppresses ²²⁰Rn gas to less than 1 % (38). Mean time for ²²²Rn 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 mm aerosol particles, the particulates from ²¹⁹Rn (half - life 3.96 s) and ²²⁰Rn (T_{1/2} 55.6 s) will be cut off and will decay while diffusing through the filter paper membrane combination. ²²²Rn (T_{1/2} 3.82 d) gas, which diffuses through the membrane, produces the alpha tracks on the detector films placed in this chamber. SSNTD placed on the other compartment having a glass fiber filter paper barrier, allows both ²²²Rn and ²²⁰Rn

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) registers 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 ⁽³⁹⁾. From the calculations by Jonsson ⁽⁴⁰⁾, it is assumed that for one hour etching at 60°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° . Upper cut-off energies, hardly changes with the angle of incidence. As a result of this, the alpha emission due to progeny (all of which have energy $> 5\text{ MeV}$) deposited on the SSNTDs are not expected to contribute tracks. This supports the assumptions made and is confirmed by experiments by placing a $^{241}\text{Am} - ^{239}\text{Pu}$ 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 undegraded 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 % ⁽⁴⁰⁾. 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 study the effect of dust load on bare card exposure mode, have shown 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 ⁽⁴¹⁾.

These dosimeters were deployed into the field on a quarterly cycle of 3 months covering all the seasons. In all about 1800 houses of different construction types spread over 45 locations in this 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 ⁽⁴²⁾.

RESULTS AND DISCUSSION

Measured ^{220}Rn gas varied from 5.7 to 42.4 Bq.m^{-3} with a GM of 12.2 Bq.m^{-3} (GSD 3.22). High ^{220}Rn levels are recorded in locations where the ^{232}Th content in the surrounding soil is high. ^{232}Th in India soil varied from 3.5 to 24.7 Bq.kg^{-1} with a mean of 18.4 Bq.kg^{-1} . ^{232}Th levels in the soil were high in the northern parts of the country ⁽⁴³⁾. ^{232}Th content in building 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 ⁽⁴⁴⁾. 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 since its formation ⁽⁴⁵⁾, which supports

the present observations. Some locations 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 survey carried out in this region shows that ^{232}Th content in the soil varied from 75 to 9070 Bq.kg^{-1} with a mean of 827.0 Bq.kg^{-1} (46), 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.

Results of a sample survey carried out in nearly 185 dwellings of different types of construction spread over four electoral wards of this 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 Bq.m^{-3} with a median value 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 value of 44.2 Bq.m^{-3} in the monazite belt region (47). Using UNSCEAR (1) dose conversion factors the

estimated annual inhalation dose due to ^{220}Rn and its progeny in Indian dwellings around normal background region varied from 0.047 to 0.39 mSv.y^{-1} with a mean of 0.14 mSv.y^{-1} (GSD 1.36).

This, when compares with the estimated inhalation dose rates of 1.05 mSv.y^{-1} for indoor ^{222}Rn and its progeny in Indian dwellings (48), total inhalation dose due to ^{220}Rn and its progeny is found to be very small. Estimated 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} , about 3.2 times higher than that recorded for the country from normal background region. Table 13 gives reported ^{220}Rn levels in dwellings and work places in literature (49, 50). ^{220}Rn levels measured in Indian dwellings are comparable with those reported for Austria, Brazil and USA. Recorded ^{220}Tn levels in dwellings in 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

Table 13. ^{220}Rn levels in dwellings in literature (Bq.m^{-3}).

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	---	---	2.0	1.0
	house	---	---	200.0	5.0
	Basement 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

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 man made, 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. Bigu⁽⁵¹⁾ reviews ^{220}Rn and ^{222}Rn progeny levels in uranium mines.

This data indicate a median ratio of PAEC(Tn) / PAEC(Rn) of 0.65 with a range of about 0.4 to over 1.5. Stranden⁽⁵²⁾ and Dixon⁽⁵³⁾, have reported measurements on a variety of underground mines and enclosures in Norway and UK. Estimated ratio of PAEC (Tn) / PAEC (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(Tn), but lower value of ratio of PAEC(Tn)/PAEC(Rn), due to generally poor ventilation of natural convection. There exists a strong correlation between PAEC (Tn) and PAEC (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⁽⁵⁴⁾ has indicated that the phenomenological relation:

$$\text{PAEC(Tn)} \propto [\text{PAEC(Rn)}]^{0.4}$$

agrees well with the indoor data on ^{220}Rn and ^{222}Rn progeny. This relationship is found to be quite consistent. Mean while the

average rate was found to be about 0.05 % or higher⁽⁵⁵⁾. 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 expected concentrations in buildings⁽⁵⁶⁾. 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⁽²³⁾. Table 14 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⁽⁵⁷⁾.

As per the regulatory body specification, the quantity of thorium allowed in a gas mantle depends on its luminous intensity. 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)^(57, 58). Besides, in the high

background areas of Chavara, Kerala, inhalation exposure due to ^{220}Rn and its progeny is also high. Table 15 gives the reported ^{220}Rn levels in some industries across the world (59-61). From this table it can

be seen the other than dwelling environment, other workplaces like gas mantle factory, monazite processing industry and thorium processing plant also have higher ^{220}Rn exposures.

Table 14. Reported $^{220}\text{Rn}/^{222}\text{Rn}$ progeny levels in literature.

LOCATION	$^{220}\text{Rn}/^{222}\text{Rn}$ Progeny	COMMENT
Italy (Latium)	1.3	Anomalous (volcanic area), 50 dwellings, poor ventilation
Canada (Elilott Lake)	0.3	Samples at 95 dwellings, source activity $^{238}\text{U}/^{232}\text{Th} \sim 1$
Hungary	0.5	22 dwellings
Norway	0.5	22 dwellings, source activity $^{238}\text{U}/^{232}\text{Th} \sim 1$
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, $^{238}\text{U}/^{232}\text{Th} \sim 0.6$
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

Table 15. Reported ^{220}Rn levels in some workplaces.

Location	No. of Data	Concentration (Bq.m ⁻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
Saktikulangara	120	7.8	56.7	14.8
K.S.Puram	125	9.0	65.7	16.9

Table 16 summarizes major findings on the data on the biological effects among humans due to exposure to thorium 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: a) 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} ; b) 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 lover dose 9.4 Gy ; ^{224}Ra emanation from the mouth greater than or equal to 24.5 Bq); and c) medical exposure to thorium oxide confining to injection of thoro-trast resulted in lifetime excess cancer risk (bone, liver) and leukemia, ranging from 55 to 330 per 10^4 persons per Gy ⁽⁶²⁾. Radionuclide of natural origin are ubiquitous in the environment at variable, but generally low, activity concentrations. The 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.

Table 16. Summary of data on biological effects due to exposure to thorium and its daughter products.

Cohort	No. of exposed persons	Exposure Characteristics	Observed effects
Non-occup. exposure: Residents in HBRA in:			
Brazil ⁽⁶¹⁾	~ 7000	^{220}Rn levels in air: $0.4 - 19 \text{ Bq.m}^{-3}$	Increased chromosome aberration
China ⁽⁶³⁾	~ 80,000	^{220}Rn levels indoors: 168 Bq.m^{-3}	Increased chromosome aberration, elevated down's syndrome
India ⁽⁶⁴⁾	~ 70,000	External dose: 7 mGy/y	Increased still birth and infant mortality. Elevated down's syndrome
Occupational exposure:			
Miners of iron ore, Rare earth ⁽⁶⁵⁾	588	Th lung burden: 0.85 Bq	Increased lung cancer incidence. Respiratory diseases
Workers in monazite industry ⁽⁶⁶⁾	300	External dose 14 mSv.y^{-1}	Increased chromosome aberrations
Workers in Th processing plant ⁽⁶⁷⁾	592	Emanating at mouth: 24.5 Bq of ^{224}Ra	Elevated SMR (lung cancer; pancreatic cancer, respiratory diseases)
Medical Exposure:			
German, Japanese and Portuguese ⁽⁶⁸⁻⁷¹⁾	~ 53,000	Bronchial life time dose: 357 mGy ; Liver dose 2.5 to 3.6 Gy/y	Liver tumors Hepatic tumors

Values of activity concentrations in materials 1 Bq.g^{-1} for uranium and thorium 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 ⁽⁷¹⁾. Table 17 gives types of operations involving naturally occurring radioactive materials pertaining to ^{232}Th , identified as required regulation on the basis of worker dose. Table 18 gives the Naturally Occurring Radioactive Material pertaining to ^{232}Th on the basis of the activity concentration reported in literature ⁽⁷²⁾.

Doses

Absorbed dose rates to the critical cells of the respiratory tract due to thoron and its progeny can be estimated on the basis of aerosol characteristics, its size distribution, unattached fraction, breathing fraction,

fractional deposition in the air ways, mucous clearance rate and location of the target cells in the airways. Most important exposure pathways is the inhalation of thoron and its daughter products. Inhaled thoron reaches equilibrium solubility only in the lung tissues, thereby causing a dose from inhaled thoron and its decay up to ^{216}Po . In case of high thoron concentration, the dose due to thoron daughter product in the lung should be considered. Such an exposure condition is possible among employees in gas mantle manufactures, where the workers closed to a pile of gas mantle house may be exposed to a ^{220}Rn concentration greater than 10, the average value of the room ⁽⁷³⁾.

Both the BEIR IV and the BEIR VI committees used dosimetric ^(62, 74). A extensive review of radon decay products activity distributions has been undertaken for occupational exposure. However, no work has been carried out so far in the case of thoron.

Table 17. Types of operations involving naturally occurring radioactive materials pertaining ^{232}Th identified as required regulation on the basis of workers dose.

Types of Operation	Naturally Occurring radioactive materials due to ^{232}Th identified as requiring regulations on the basis of workers dose			
	Description	Dominant nuclide	Conc. (Bq.g^{-1})	Workers dose (mSv y^{-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 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

Table 18. Naturally occurring radioactive material that is considered for regulation pertaining to ^{232}Th on the basis of activity concentration.

Material Category	Material	Predominant Nuclide	Typical Activity (Bq/g)
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	^{232}Th	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	^{238}U	46 – 70
	Zircon refractory	^{238}U	0.14 - 2
Slag	Niobium extraction	^{232}Th	20 to 120
	Tin smelting	^{232}Th	0.07 – 15
Scales, Sludge, sediments	BaSO_4 precipitate	^{232}Th	200

It is not possible to assess the radiation dose from inhalation of thoron decay products by means of epidemiological means and therefore it has to be evaluated using dosimetric modeling. According to UNSCEAR ^(1, 75) the dose conversion factors have been estimated taking into account ^{212}Pb from the lung. The estimated dose conversion factor varies drastically based on the breathing rate as well as the target tissue mass. Lung dose distribution assessment carried out by different agencies from 1956 to 2000 shows that the dose conversion factors estimated varied from 30 to 103 nSv/Bq $\text{h}\cdot\text{m}^{-3}$ (table 19) UNSCEAR ⁽²³⁾. This table supports the continuous use of the conversion factor of 40 nSV/Bq $\text{h}\cdot\text{m}^{-3}$ for thoron progeny indoors and outdoor used for the estimation of inhalation dose due to thoron and its progeny as given in table 20 ⁽¹⁾. In this table EEC is the equilibrium equivalent concentration of thoron and is the product of the concentration of thoron and the equilibrium factor between thoron and its decay products. The equilibrium factor is taken as 0.1 for outdoor and 0.03 for indoor. These values are weighted for an occupancy factor of 0.2 for outdoor and 0.8 for indoor.

Among residents in high background areas in Brazil, China and India (total population: ~ 157,000 persons), statistically significant increases have been observed for chromosome aberration and down's syndrome. Only the Chinese study addressed the lung cancer issues, and found no increase in lung cancer (mean ^{220}Rn level indoors 168 Bq. m^{-3}), but an increase in respiratory diseases ⁽⁶³⁾. Among occupationally exposed persons (iron and rare earth miners, workers in Th-processing plant and monazite industry; total work force; 1557 persons), respiratory diseases, pancreatic cancer, and chromosome aberrations were found to be statically significantly increased). Among niobium miners in a Th-rich area, a significant increment in lung cancer (ratio observed/ expected lung cancer cases = 11.3) was also detected ⁽⁷²⁾. European and Japanese patients (total number = ~ 53,000 persons), treated with thorium oxide injections, have experienced a statistically significant life time excess cancer risk (liver to bone) and leukemia. However, no excess lung cancer was noticed, despite an average bronchial life time dose of 357 mGy.

Table 19. Principle dosimetric assessments of lung dose from the deposited thoron decay products since 1956 till 2006 ⁽²³⁾.

Year	Parametric value		Target region	Model type	Effective dose (nSv/Bqh m ⁻³)
	Unattached fraction (²¹² Pb)	Breathing rate (m ³ h ⁻¹)			
1956	0.02	0.3	Average in 45 m epithelium	Cast of Trachea and bronchi	30
1959	0.02	1.2	Mean TB region	Deposition retention assumption	43
1973	0.02	0.9	Basal cell generations 2 -15	weibel	35
1980	1.2	-do-	Bronchial from lung x 0.06	64
1981	1.2	-do-	Based on Jacobi and Einfeld	73
1982	1.2	Mean bronchial	-do-	34-103
1983	0.02	1.2	-do-	- do -	36
2000	1.2	Whole body	ICRP- 50	40
2001	0.005 – 0.02	0.75	Bronchial	Modified from ICRP-66 weighing by basal and secretory cell density	53

Table 20. Average concentration of thoron and its progeny in air and corresponding annual effective doses.

Nuclide	Location	Concentration (Bq m ⁻³)		Effective dose equivalent (nSv/Bq h m ⁻³)		Annual effective dose (m Sv)	
		Gas	EEC	Gas	EEC	Gas	EEC
Thoron	outdoor	10	0.1	0.11	40	2.0	7.0
	indoor	10	0.3	0.11	40	8.0	84
Total							101

CONCLUSION

²²⁰Rn levels in Indian dwellings varied from 5.7 to 42.2 Bq.m⁻³ with a GM of 12.2 Bq.m⁻³. Higher ²²⁰Rn levels are recorded in dwellings around locations where the ²³²Th content in soil also high. Estimated national average value of ²²⁰Rn levels for India are comparable with those reported for Austria,

Brazil and USA. Inhalation dose rate due to ²²⁰Rn and its progeny varied from 0.047 to 0.39 mSv/y with a GM of 0.14 mSv/y. Inhalation dose rate due to ²²⁰Rn 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. Estimated ration of ²²⁰Rn/²²²Rn 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. Surveys carried out in some western countries are indicative of a non negligible $^{220}\text{Rn} / ^{222}\text{Rn}$ exposure component for some members of the general public. This is also the case with occupational exposure received at work places, particularly in monazite processing industry. But the presentably available data on $^{220}\text{Rn} / ^{222}\text{Rn}$ daughter levels, aerosol characteristics, and their behavior outdoors and indoors cannot be considered as representative one.

There is also ambiguity in the current understanding of potential health deterrents due to the lack of any established dose effect relationship and contradictory evidence of biological effects induced by $^{220}\text{Rn} / ^{222}\text{Rn}$ daughters. So efforts should be warranted to address the $^{220}\text{Rn} / ^{222}\text{Rn}$ daughter issue on an international scale. Such a coordinated research programme should include industrialized countries as well as developing nations, since both seems to be affected. Emphasis should be on areas which will assist in improving the current dose assessment of population groups estimated to receive partly significantly elevated $^{220}\text{Rn} / ^{222}\text{Rn}$ daughter exposures.

In summary, our scientific knowledge is such that, on average, world wide ^{220}Rn is expected to be less of a problem than ^{222}Rn . In view of limited resources, research work should focus 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 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 $^{222}\text{Rn} + ^{220}\text{Rn}$; ^{220}Rn can thus be a source of error in residential ^{222}Rn studies which do not distinguish the two contributions to

exposure^(71,72). Further measurement studies are needed to consider the contribution of both ^{222}Rn and ^{220}Rn .

In view of the generally weak international data base, representative data base are needed on the following topic areas so as to assess the significance of the $^{220}\text{Rn} / ^{220}\text{Rn}$ -d issues:

1. Radon-220 contribution from different source-terms in domestic and occupational exposure situations
2. Laboratory and field tests of $^{220}\text{Rn} / ^{220}\text{Rn}$ -d measurement and monitoring equipments
3. Development of inexpensive and user-friendly, convenient equipments
4. QA/QC procedures concerning the meteorology of thoron /thoron progeny for research and radiation protection purposes
5. Simultaneous measurement of thoron/thoron daughter outdoors and indoors deposition, attachment, and to determine particle size distribution, disequilibrium, and air exchange processes in various environment
6. Radon-220 exhalation rates for most frequently used building materials and soil types, and its modification by environmental and meteorological parameters
7. Radon-220 /radon-220 daughter domestic exposure levels
8. Radon-220/radon-220 daughter occupational exposure levels
9. Domestic parameters for $^{220}\text{Rn} / ^{220}\text{Rn}$ -d inhalation, including lung deposition, clearance, and retention processes
10. Inhalation studies with realistic $^{220}\text{Rn} / ^{220}\text{Rn}$ -d concentration levels similar to occupational exposure situations to corroborate internal dosimetry modeling.

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