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ORIGINAL ARTICLE

Measurements of radon (^{222}Rn) and thoron (^{220}Rn) exhalations and their decay product concentrations at Indian Stations in Antarctica

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Abstract
During the 33rd summer expedition to the two Indian Stations at Antarctica, Bharati and Maitri, radon and thoron progeny concentrations in indoors and outdoors were measured along with radon (^{222}Rn)/thoron (^{220}Rn) exhalation rate measurements of soil samples, radionuclide content and ^{222}Rn emanation coefficient. This investigation was based on the reports of the higher gamma radiation levels reported around these stations. The results will give an estimate of the radioactivity level as well as the total dose received by personnel carrying out long-term measurements at these stations. Radon and thoron progeny concentrations were measured using direct radon and thoron progeny sensors (DRPS and DTPS). The soil radionuclide content was measured using HPGe gamma spectrometry while radon/thoron exhalation rates were measured using the accumulation method by scintillation radon/thoron monitor. In contrast to a higher radiation field and radioactivity content, studies showed the radon/thoron exhalation rates and progeny concentration to be similar to that measured in normal background areas of other parts of the world. This could be attributed to the ice deposits and the larger atmospheric dispersion, and also to the soil nature which is mainly loamy sands with low clay content contributing to a lower emanation. The results are discussed.

Keywords Antarctica · Radionuclide content · ^{222}Rn/^{220}Rn exhalation rate · EETC · EERC · Accumulation method · DTPS/DRPS

Introduction

Antarctica has ice coverage of 98% and possible larger deposits of Uranium and Thorium in sedimentary basins that existed prior to the breakup of Gondwana. Uranium minerals or anomalous levels of radioactivity have been reported in several locations in Antarctica, particularly in Enderby Land, the Adelie Coast and the Trans-Antarctic Mountains of Victoria Land in East Antarctica (Congress 1989). Radon (^{222}Rn) and its isotope thoron (^{220}Rn) are naturally occurring radioactive gases, continuously being generated in rocks and soil containing primordial radionuclides uranium and thorium, respectively. The process by which radon and thoron escapes from the rock/soil matrix and reaches to the atmosphere is called exhalation (Nazaroff and Nero 1988). ^{222}Rn exhalation from a matrix is mainly governed by coefficient of radon emanation from soil grain to the porous volume and then diffusion rate in pore space of the matrix (Sahoo and Mayya 2010). ^{222}Rn emanation coefficient is generally higher for fine soil grain and lesser for coarse grain. ^{222}Rn diffusion in the pore space depends upon the porosity and moisture content in the soil matrix. For example, clay loam soils are characterised by its high sorptivity or low infiltration capacity (Dutt et al. 2012; Verma et al. 2015; Maheshwar et al. 2015; Szajdak and Karabanov 2010), and hence may hold more water content in the pores, enabling trapping of more ^{222}Rn atoms within the matrix and thereby diffusing lesser ^{222}Rn atoms to the atmosphere.

Jonas et al. (2016) had carried out thoron exhalation of the samples and observed that it increased as a function of the Th-232 content and the emanation factor as well. Further, no correlation was found between the Th-232 content and the emanation factor. The results clearly proved that the emanation factor is of great importance with regard to exhalation

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since in spite of high Th-232 content, the measured thoron exhalation of the samples was low. They had also compared thoron emanation- and exhalation-related results with ^{222}Rn emanation and exhalation results investigated on the same soil samples previously by Kardos et al. (2015), and found no correlation in spite of the relatively similar recoil energies of the decay of their parent elements. Masahiro et al. (2008) in their radon and thoron exhalation rate studies in Japan observed that the exhalation rate increased in proportion to the SiO_2 concentration in the soil and that the average values were 8.6 mBqm^{-2}s^{-1} and 0.80 Bqm^{-2}s^{-1}, respectively. Further, it was concluded that the thoron exhalation rate was about 100 times of the radon exhalation rate. Goto et al. (2008) had developed a radon exhalation rate distribution model, which considers dependency on soil water saturation, soil temperature and soil texture. Using this model, they had given the global average of the ^{222}Rn exhalation rates to be ~ 18 mBqm^{-2}s^{-1} which was slightly less than widely used values such as 20.8 mBqm^{-2}s^{-1}.

^{222}Rn and ^{220}Rn exhalations from earth surface are considered as the primary source term for atmospheric ^{222}Rn and ^{220}Rn. ^{222}Rn/^{220}Rn exhalation data provide additional supportive information towards detection of any elevated uranium/thorium content in rocks as well as a precursor for decay product concentrations which generally contribute about 50% of natural background radiation dose (UNSCEAR 2000). ^{222}Rn studies could offer some important information pertaining to global climatic changes (Park 1993; Rusov et al. 2003). Various ^{222}Rn measurements have been reported by several stations at Antarctica to being about two to three orders of magnitude less than in other continents with values varying from 0.01 to 0.03 Bqm^{-3} (Ilic et al. 2005; Hirotaka et al. 1998; Pereira et al. 1988).

National Centre for Antarctic and Ocean Research (NCAOR), India, has established two research stations in Antartica, one is the Maitri Station in the Schirmacher Oasis (70°45′S:11°45′E), East Antarctica; and the second is Bharati in the Larsemann Hills between Thala Fjord & Quiltybay, east of Stornes Peninsula in Antarctica at 69°24.41′S, 76°11.72′E, approximately at 35 m above sea level (Fig. 1). Adhering to Environmental protection protocol by the Antarctic Treaty which includes India, environmental assessment exercise covering soil and air with respect to radiological parameters has been studied by Bhabha Atomic Reasearch Centre (BARC), India. Under this program, the different studies carried out include the measurement of cosmic ray dose, gamma dose rate, the radionuclide contents of the Antarctic soil and the measurement of ^{222}Rn and its decay products as these are the primary contributors to the natural radiation dose.

The highlights of the earlier expeditions at the two stations are as follows:

At Indian Station Maitri, the gamma radiation levels were measured to be in the range of 100–200 nGy/h during the 32nd expedition program (Bakshi et al. 2013). Earlier during 10th expedition program, Jojo et al. (1995)

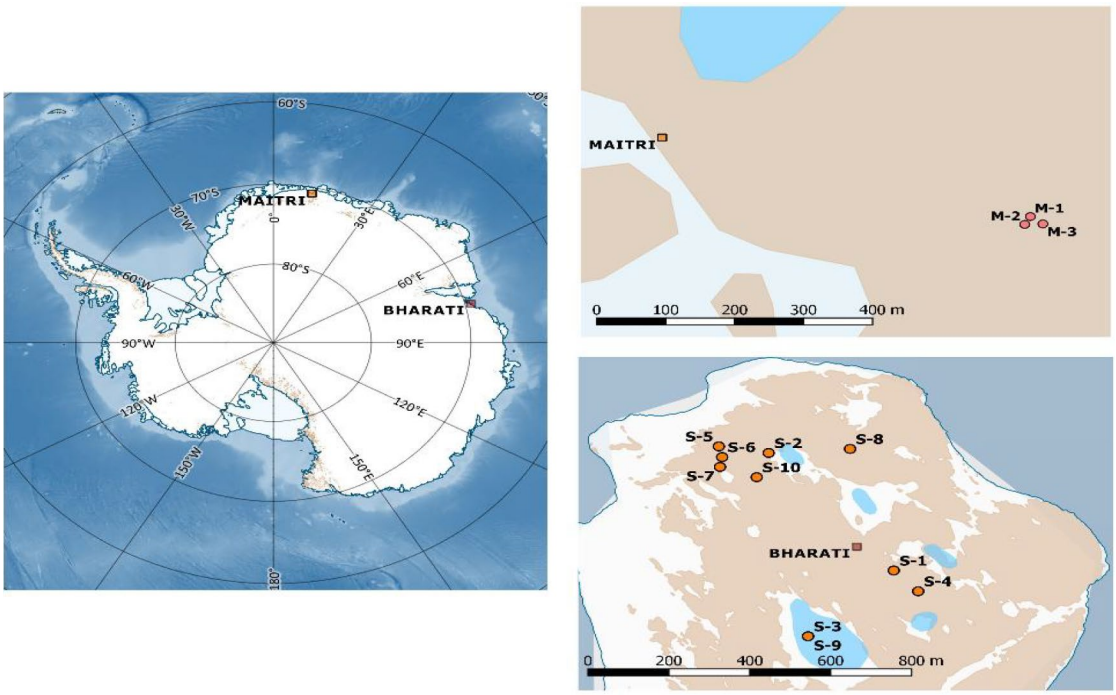

Fig. 1 Map of Antarctica with the sampling locations at Bharati and Maitri stations

had measured the uranium content in the soil by fission track analysis method and reported its value varying from 0.45 to 4.49 $BqKg^{-1}$. In the same expedition program, measurement of ^{222}Rn and its decay products were also carried out. Activity concentration of radon in the outdoor air measured using grab filter-paper sampling and alpha counting was found to be varying from 0.02 to 0.03 Bqm^{-3}. Ramachandran and Balani (1995) have reported the ^{222}Rn decay product levels indoors (inside the station) measured using bare LR115-type SSNTD, to be varying from 1.7 to 21.3 Bq m^{-3}. This bare-detector has a lot of interference from radon and thoron gas also; hence, it has now been discarded as a measurement technique. In addition, the authors had also mentioned that the higher value of 21.3 Bqm^{-3} was obtained from the room which remained always closed and is not used for staying. Also, ^{220}Rn was being neglected owing to its shorter half-life (55 s) but recently reports have shown significant radiological importance of ^{220}Rn considering its harmful health effects (Tokonomi et al. 2008); to overcome this disadvantage, in the present work, it was decided to carry out some additional measurements in Maitri station using the recently developed direct radon and thoron progeny sensors (Mishra et al. 2014). In the present work, measurements were carried out in the rooms where people were staying so as to estimate the inhalation dose.

Also at Bharati Station, gamma radiation levels in the range of 600–700 nGy/h have been reported (Bakshi et al. 2013), which is a few times higher than that measured in Maitri Station. In this line, radioactivity measurements were carried out in soil samples collected from Bharati and Maitri Stations (Bakshi et al. 2016). The measured high radium(^{226}Ra), thorium(^{232}Th) and potassium contents(^{40}K) in Bharati samples compared to Maitri samples are in conjunction with the reported high gamma dose rate in Bharati region (Bakshi et al. 2013). Since ^{222}Rn and ^{220}Rn originates from ^{226}Ra and ^{232}Th, respectively, the exhalation rate of ^{222}Rn and ^{220}Rn from these regions could be high and so also the atmospheric $^{222}Rn/^{220}Rn$ decay product concentration. In an earlier study carried out by Jonas et al. 2016, it has been reported that no correlation was found between the Th-232 content and the emanation factor, and that in spite of high Th-232 content, the measured thoron exhalation could be less. So, this intercomparison study between the two stations and correlation between different radon and thoron related soil parameter study was taken up during the 33rd Indian Scientific Expedition to Antarctica in the period of November–March, 2013–2014, wherein the following studies were aimed at: measurement of (1) soil radon (^{222}Rn)/thoron (^{220}Rn) exhalation rate measurements, (2) radionuclide content in the soil, (3) radon emanation coefficient from soil samples, (4) radon(^{222}Rn)/thoron (^{220}Rn) progeny measurements in indoors and outdoors of the stations using Solid State Nuclear Track Detectors (SSNTD) based DTPS/DRPS systems.

Materials and methods

Soil sample collection

Ten soil samples from Bharati Station and three from Maitri Station were collected during November–March 2013–2014 from the undisturbed areas from the different locations determined based on the gamma radiation levels measured using a GM-based Survey meter and processed as per the IAEA Protocol (IAEA TRS-295). In all the locations, a 1 m × 1 m area was demarcated and the area was cleared of stones, pebbles and other organic matter. Around 1.5 kg of soil samples was collected at a depth of 15 cm, packed and stored in a sealed polyethylene bags marked with GPS details and survey meter readings. The raw soil samples in the same form as collected (with the same moisture content, grain size, temperature, etc.) were first subjected to $^{222}Rn/^{220}Rn$ exhalation studies and then analyzed for radioactivity content which is used for the estimation of emanation coefficient. The samples after collections were kept in air tight plastic bags so that soil moisture would not change much. It has been reported that for sandy loam type of soils prevailing in Antarctica during summer months H_2O in situ % is around 1.5 at a depth of 2–10 cm and for surface 2 cm it is 5.0. Similarly, the temperature at 10 cm in soil is around 0 °C and at 2.5 cm 4 °C, and surface in sun is 12 °C (Cameron et al. 1970).

Measurement of thoron exhalation rate

Due to the short radioactive half-life of ^{220}Rn (55.6 s), the diffusion length of ^{220}Rn is only about 1 cm. This means that only the top surface contributes to the ^{220}Rn exhaling to the environment and hence the exhalation rate is called the surface exhalation rate. Also by changing the geometry of the accumulator, it can be shown that ^{220}Rn mass exhalation rate may change but surface exhalation rate (normalised with respect to surface area) will not change. The active measurement of ^{220}Rn surface exhalation rate was carried out by hermetically sealing the soil samples in a cylindrical chamber of inner diameter 120 mm and height 120 mm. This was followed by measuring the equilibrium ^{220}Rn concentration by ZnS(Ag) scintillation-based Continuous Thoron monitor, namely Smart Thoron Monitor, STM (Gaware et al. 2011), as depicted in the experimental setup shown in Fig. 2. The chamber was connected to STM through an air flow pump (flow rate = 1 l min^{-1}) forming a closed circuit loop so that the ^{220}Rn gas which builds up in the empty volume of the cylindrical chamber enters the detector volume of the STM. The rate of change

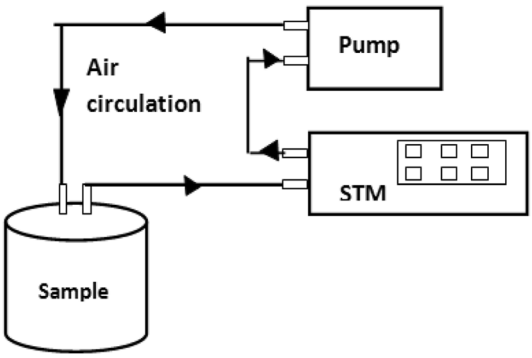

Fig. 2 Schematic diagram for measurement of ^{220}Rn exhalation from sand sample using STM in flow mode of sampling

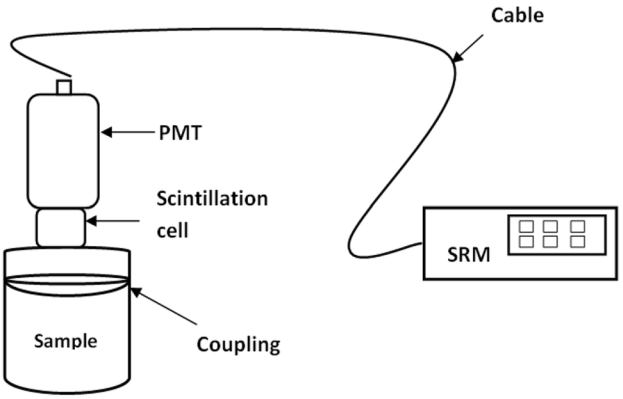

Fig. 3 Schematic diagram for measurement of radon exhalation rate from sand samples using SRM in diffusion mode of sampling

of ^{220}Rn activity concentration, C(Bq m^{-3}), in the closed loop is given by Kanse et al. (2013):

$$\frac{dC}{dt} = J_T A - CV'\lambda \tag{1}$$

where J_T is the surface exhalation rate of ^{220}Rn from the soil in (Bq m^{-2} s^{-1}), V' the volume enclosed in the closed loop in m^3 (i.e., sum of residual volume of chamber, tube volume and detector volume), λ the decay constant of ^{220}Rn (0.012464 s^{-1}) and A is the area of cross section of the chamber. At equilibrium, the ^{220}Rn exhalation rate is given by the following expression (Sahoo et al. 2014):

$$J_T = \frac{CV'\lambda}{A} \tag{2}$$

The detection limit of STM is 12 Bqm^{-3} at 1 h counting cycle.

Measurement of radon exhalation rate

For measurement of radon exhalation rate, the same cylindrical accumulation chamber filled with the soil sample which was used for thoron exhalation rate measurement is now connected to a continuous radon monitor, Smart Radon Monitor, SRM (Gaware et al. 2011), in which the buildup of ^{222}Rn gas in the chamber was monitored. In case of ^{222}Rn, the diffusion length in soil is of the order of 1 m due to its longer half-life (3.82 days) and hence the total mass of the sample contributes to the exhalation process, and hence the exhalation rate measured here is the mass exhalation rate. The schematic diagram of the experimental set up is shown in Fig. 3. ^{222}Rn concentration which builds up with time in a closed chamber is described by the following linear form (Sahoo et al. 2007):

$$C(t) = \frac{J_m M}{V} t + C_0 \tag{3}$$

where $C(t)$ is the ^{222}Rn activity concentration at time t in the chamber volume (Bq m^{-3}); C_0 the ^{222}Rn concentration present in the chamber volume at $t=0$ (Bq m^{-3}), J_m the mass exhalation rate of ^{222}Rn from the sample (Bq kg^{-1} h^{-1}), M the total mass of the sample (kg), t the measurement time (h) and V is the effective air volume of the set-up which is the sum of the residual volume of the chamber, porous volume (V_P) of the sample and detector cell volume.

The porous volume of the sample is defined by the following equation:

$$V_P = V_S - \frac{M}{\rho_g} \tag{4}$$

where V_S is the sample volume (m^3), M the mass of the sample (kg) and ρ_g is the soil specific gravity which is 2700 kg m^{-3} (UNSCEAR 2000; Prakash et al. 2012).

Using Eq. 3, ^{222}Rn mass exhalation rate can be estimated as

$$J_m = \frac{B.V}{M} \tag{5}$$

where B is slope obtained from the linear fit to the plot of ^{222}Rn concentration versus time. Then from the obtained mass exhalation rate and radium content of the sample, ^{222}Rn emanation fraction is calculated as

$$f = \frac{J_M}{Q\lambda} \tag{6}$$

where Q is the radium content in Bq kg^{-1}, obtained from HPGe Gamma spectrometer.

The detection limit of SRM is 8 Bqm^{-3} of radon at 1-h counting cycle.

Measurement of ^{226}Ra, ^{232}Th and ^{40}K contents in soil samples

After carrying out the exhalation studies, the soil samples were sieved and oven dried at 110 °C for 24 h to remove the

moisture. The samples were then filled in the plastic containers having dimension and geometry (6.2 cm height and 5.8 cm diameter) approximately same as that of the IAEA standard Sources, RGU, RGTh and RGK used for efficiency calibration. These samples were sealed using Araldite gum for making it radon impermeable and left for 28 days so as to bring ^{222}Rn and its short-lived decay products into secular equilibrium with ^{226}Ra (Mauring and Gäfvert 2013).

The measurement of ^{226}Ra, ^{232}Th and ^{40}K contents of the samples was carried out in P-type Coaxial HPGe gamma radiation detection unit, GCD-30 185 with 30% relative efficiency in horizontal cryostat. The energy and efficiency calibration of the system corresponding to a particular radionuclide/gamma line was determined using the IAEA certified standard source RGU, RGTh and RGK.

The background corrected spectrums of the samples were obtained and the activity of radium, thorium and potassium was estimated using the following expression (IAEA 1989a, b):

$$A = \frac{N * 100 * 100}{\eta * p * W} \tag{7}$$

where N is the net count rate under the photo-peak for a given gamma line under study, A the specific activity of the radionuclide in Bq kg^{-1}, P the percentage gamma abundance of a particular gamma line, η the photo peak efficiency of the detector system and W is the weight of the sample (kg). For the estimation of radium, energy peaks of 186.1 keV from ^{226}Ra, 609.4 and 1764.5 keV from ^{214}Bi were used while for ^{232}Th estimation, 911 keV from ^{228}Ac, 238.6 keV from ^{212}Pb, 583.1 and 2614.5 keV from ^{208}Tl were used. For ^{40}K estimation, the photo peak energy of 1.46 MeV emitted by potassium itself was used (IAEA 1989a, b). The overall error in estimation was calculated to be around ± 15%. The MDA (Minimum Detectable Activity) of HPGe detector system for ^{40}K, ^{226}Ra and ^{232}Th were 16.6 Bq kg^{-1}, 6.7 Bq kg^{-1} and 7.6 Bq kg^{-1} respectively, for a counting time of 100,000 s and 400 g weight of sample.

Estimation of ^{226}Ra equivalent and external dose rate

To represent the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, a common radiological index, called radium equivalent activity ($^{226}Ra_{eq}$) was estimated, which is given by the expression (UNSCEAR 2000),

$$Ra_{eq} = 1.43C_{Th} + C_{Ra} + 0.77C_{K} \tag{8}$$

where C_{Th}, C_{Ra} and C_K are the activity concentrations (in Bq kg^{-1}) of ^{232}Th, ^{226}Ra and ^{40}K, respectively. It was assumed that ^{226}Ra is in equilibrium with ^{238}U. The radiation dose rate, D (nGy h^{-1}), expected to be present at the sampling location at about 1 m height resulting from the natural radio-nuclides in the soil was computed using the dose conversion co-efficient given by (UNSCEAR 2000) using the following equation:

$$D = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_{K} \tag{9}$$

where C_{Ra}, C_{Th} and C_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in soil samples (in Bq kg^{-1}).

^{222}Rn/^{220}Rn progeny measurements

^{222}Rn/^{220}Rn progeny measurements were carried out in eight indoor locations at the Maitri Station. For this measurement of ^{222}Rn and ^{220}Rn progeny, Solid State Nuclear Track (SSNTD) based Direct Radon Progeny Sensors (DRPS) and Direct Thoron Progeny Sensors (DTPS) (Mishra and Mayya 2008, 2009a) were used. The advantage of these detectors is that the tracks due to the alpha particles emitted from ^{222}Rn and ^{220}Rn progeny are permanently registered in these SSNTD films which can be counted in a later convenient time for the estimation of progeny concentration. These sensors were suspended at a distance of 30 cm from the walls and exposed for a period of 4 months. Simultaneously, active measurements of ^{222}Rn/^{220}Rn progeny activity concentrations were carried out at five outdoor locations of Maitri station using flow mode DRPS and DTPS loaded integrated samplers (Mishra et al. 2009b) kept at a distance of 1.5 m from the ground. Integrated sampler consists of a filter paper and a detector (DRPS/DRTPS) arrangement in the sample holder attached to a pump operating at a flow rate of 2 lpm. Air is sampled such that the ^{222}Rn and ^{220}Rn progeny are deposited on the filter paper; the DTPS/DRPS placed facing the filter-paper registers the alpha tracks emitted from the decay of the deposited activity. The calibration of these detector systems is explained elsewhere (Mishra et al. 2009a, b). The experimental set-up for the measurement of ^{222}Rn/^{220}Rn progeny concentrations in outdoor location of Maitri Station using flow mode integrated sampler is shown in Fig. 4. The sampling was carried out for ~ 100 h. After the exposure, the detectors were retrieved and were analyzed afterwards in the laboratory. The track densities in these detectors were estimated following chemical etching using a standard protocol of 2.5N NaOH at 60 °C for 90 min. The track densities were related to ^{222}Rn and ^{220}Rn progeny concentrations using appropriate calibration factors (Mishra et al. 2009b). From these measured progeny concentrations, inhalation dose, D (nSv), could be established using the following relation:

$$D = (9 * \mathrm{EECR} + 40 * \mathrm{EECT}) * \mathrm{ET} \tag{10}$$

where EECR and EETC are the ^{222}Rn and ^{220}Rn progeny concentrations (Bq m^{-3}), 9 and 40 are the dose conversion factors (nSvh^{-1} per Bq m^{-3}) for ^{222}Rn and ^{220}Rn progeny, respectively, as proposed by UNSCEAR (2000), and ET is

Fig. 4 Experimental set-up for the measurement of ^{222}Rn/^{220}Rn progeny concentrations in outdoor location of Maitri station using flow mode integrated sampler

the exposure time (h). The detection limit for DTPS system is 0.1 Bqm^{-3} and for DRPS system is 1 Bqm^{-3}.

Results and discussion

^{220}Rn and ^{222}Rn exhalations from soil samples

^{220}Rn and ^{222}Rn exhalation rates and ^{222}Rn emanation fraction of the soil samples obtained using Eqs. 2, 5 and 6 are listed in Table 1. The average ^{222}Rn mass exhalation rates obtained from Bharati and Maitri samples are (5.17 ± 1.92) mBq kg^{-1} h^{-1} and (2.81 ± 0.37) mBq kg^{-1} h^{-1}, respectively, which show that ^{222}Rn mass exhalation rate in Bharati samples was about two times higher than the Maitri. This is because of the increased radium content in Bharati region compared to Maitri. The measured mass exhalation rates are within the reported range of 2.8–63 mBq kg^{-1} h^{-1}(Hussein et al. 2013a, b; Kobeissi et al. 2008; Kumar et al. 2015; Kumar and Singh 2004; Shoeib and Thabayneh 2014; Rafat 2015; Sahoo et al. 2007). The average value of ^{222}Rn emanation fraction obtained from Bharati and Maitri samples are (1.81 ± 0.26)% and (2.72 ± 0.41)%, respectively. These are relatively low compared to that reported for soil samples from several countries (Nazaroff et al. 1989; Stoulos et al. 2003, 2004; Sahoo et al. 2007). Stoulos et al. (2004) measured the radon emanation faction from different building materials used in Greece and reported its value varying from 1.6% in Pozzolan Portland Cement to 25% in soil. Sahoo et al. (2007) measured radon emanation fraction in the Indian building materials and reported

Table 1 ^{222}Rn mass exhalation and ^{220}Rn surface exhalation rates from the sand samples from Antarctica (B for Bharati Station and M for Maitri Station)

Sample no.	GPS data	^{226}Ra content (Bq kg^{-1})	^{222}Rn Mass exhalation rate (mBq kg^{-1} h^{-1})	^{222}Rn emanation fraction (%)	^{232}Th content (Bq kg^{-1})	^{220}Rn surface exhalation rate (Bq m^{-2} h^{-1})
B-1	69°24′26.7"S 76°11′22.5"E	54.24 ± 1.3	7.12 ± 0.81	1.74 ± 0.20	261 ± 2.4	1111 ± 76
B-2	69°24′33.6"S 76°10′43.5"E	24.8 ± 1.9	3.20 ± 0.50	1.72 ± 30	138.3 ± 4.5	557 ± 42
B-3	69°24′34.9"S 76°11′35.9"E	40.6 ± 2.8	6.86 ± 1.26	2.25 ± 0.44	214.9 ± 3.2	1075 ± 46
B-4	69°24′25.3"S 76°11′29.5"E	28.4 ± 1.9	3.64 ± 0.52	1.71 ± 0.27	192.1 ± 3.1	1092 ± 85
B-5	69°24′37.3"S 76°10′39.0"E	46.1 ± 1.8	7.86 ± 1.56	2.26 ± 0.46	250 ± 2.9	1086 ± 30
B-6	69°24′37.3"S 76°10′42.1"E	35.52 ± 1.5	4.57 ± 1.02	1.7 ± 0.39	219 ± 2.4	1148 ± 112
B-7	69°24′37.3"S 76°10′44.7"E	25.2 ± 1.9	3.35 ± 0.78	1.77 ± 0.43	187.9 ± 3.2	992 ± 54
B-8	69°24′27.2"S 76°10′46.8"E	32.9 ± 2.0	4.22 ± 0.75	1.71 ± 0.32	268.4 ± 2.9	1135 ± 70
B-9	69°24′34.9"S 76°11′35.9"E	33 ± 1.5	3.44 ± 0.42	1.38 ± 0.17	183 ± 2.3	887 ± 25
B-10	69°24′34.9"S 76°11′35.9"E	53.3 ± 2.2	7.51 ± 1.35	1.87 ± 0.34	324.7 ± 3.7	1195 ± 107
M-1	70°45′53"S 11°44′57"E	18.18 ± 1.5	3.15 ± 0.45	2.3 ± 0.38	32 ± 2.0	123.5 ± 11
M-2	70°45′53.5"S 11°44′56.5"E	10.27 ± 1.3	2.41 ± 0.5	3.1 ± 0.50	34.33 ± 2.0	155.1 ± 10
M-3	70°45′53.3"S 11°44′59"E	13.71 ± 1.5	2.86 ± 0.5	2.75 ± 0.57	31.59 ± 2.2	107.9 ± 10

its value ranging from 0.1% in vitrified tiles to about 20% in soil and lime. Nazaroff et al. (1989) have reported the radon emanation coefficients for soils of 13–20% of dry weight to be ranging from 22 to 32%, for moist soil to be ranging from 3 to 55% and for sandy loam to be ranging from 10 to 36%.

The lower ^{222}Rn exhalation rate from the samples may be attributed to the larger grain size of samples collected from Antarctica compared to normal soil. Moreover, it has been reported that the Larsemann Hills, wherein Bharati station is located, is an ice-free hilly terrain and the gravelly sand present there consists of low clay content ranging from 2 to 8% (Mergelov 2014). Also, the soils prevalent in these areas are mainly sands and loamy sands with larger grain size having lesser specific surface area (m^2/gm) for appreciable ^{222}Rn emanation to occur henceforth a lower radon emanation coefficient, which is revealed in this study. The relatively low clay contributes to a low value of emanation coefficient since for high silt and clay content, considerable ^{222}Rn escapes the grains via microcracks and defects (Greeman et al. 1996; Schery et al. 1984).

Also in situ ^{222}Rn exhalation is site as well as time specific. It depends mainly upon radium content, bulk density and diffusion coefficient of soil at the site and varies with time depending upon the moisture content and ambient temperature in soil. Hence, it is a cumbersome task to perform several measurements on-site at different times of a day or in different seasons to evaluate the representative radon exhalation potential at the site. In contrast, ^{222}Rn mass exhalation rate of the dry soil sample is a stable parameter and widely used for characterizing and comparing the radon exhalation potential of different matrices. Once this quantity is measured for the sample collected without any alteration in grain size, then it eliminates the dependency of radon exhalation on radium content and grain size. With this parameter, representative radon exhalation potential of the site can be evaluated easily by knowing bulk density and diffusion coefficient of soil, and variation in moisture content and ambient temperature in soil using radon diffusion and advection models (IAEA TRS 474 2013; Sahoo and Mayya 2010; Amit; Kumar et al. 2014; Chakraverty et al. 2018).

The average ^{220}Rn surface exhalation rate obtained from Bharati and Maitri samples are (1028 ± 186) Bq m^{-2} $h^{-1}$and (128.8 ± 24) Bq m^{-2} h^{-1}, respectively. Thoron exhalation rate from Bharati samples are about five to ten times higher than the Maitri samples which could be attributed to higher thorium content in Bharati samples. But all these values for thoron exhalation rate are much lesser than the global average which is 3600 Bq m^{-2} h^{-1} as reported by UNSCEAR (2000). Also in case of thoron surface exhalation rate, there is no much variation expected between in situ and ex situ values because only the top surface layer of the soil contributes to the exhalation owing to its short diffusion length (~ 1 cm). Moreover, the samples were packed in air tight plastic containers so as to maintain the soil parameters such as moisture content.

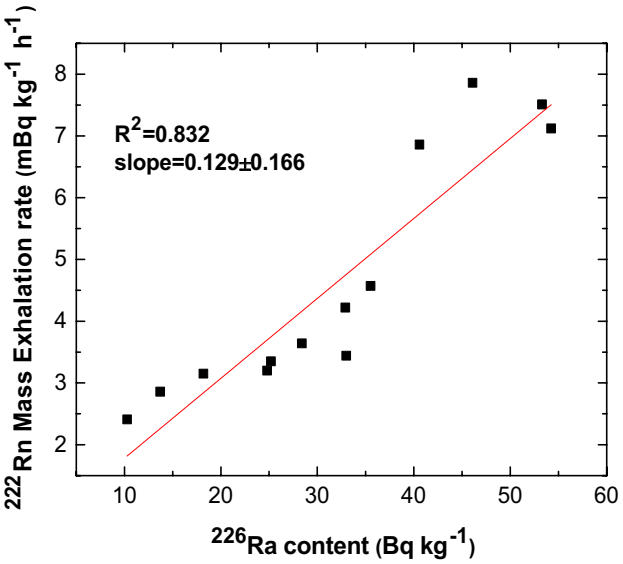

Fig. 5 Linear correlation analysis between ^{226}Ra content and ^{222}Rn mass exhalation rate of soil samples collected from Antarctica

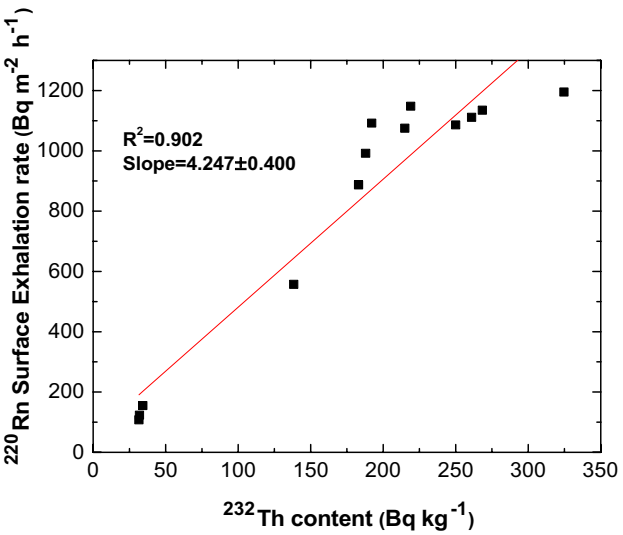

Fig. 6 Linear correlation analysis between ^{232}Th content and ^{220}Rn surface exhalation rate of sand samples collected from Antarctica

A correlation study of ^{222}Rn exhalation rate with respect to ^{226}Ra content, as shown in Fig. 5, showed a good linear correlation. A similar correlation of ^{220}Rn exhalation rate with ^{232}Th content is shown in Fig. 6, which also shows a good correlation between the two quantities.

Radionuclide content in soil samples

The activity concentration of the radionuclide ^{226}Ra, ^{232}Th and ^{40}K and the estimated radium equivalent and dose rate at 1 m from ground for the soil samples collected from Bharati and Maitri stations, Antarctica, are listed in Table 2. The mean (±SD) of ^{226}Ra, ^{232}Th and ^{40}K contents of the Bharati samples were obtained as 37.42 ± 10.8 Bq kg^{-1}, 224.12 ± 53.4 Bq kg^{-1} and 996.05 ± 131.8 Bq kg^{-1}, whereas for Maitri samples the mean (±SD) values of ^{226}Ra, ^{232}Th and ^{40}K contents were obtained as 15 ± 3.4 Bq kg^{-1}, 32 ± 1.5 Bqkg^{-1} and 522.28 ± 39.3 Bqkg^{-1}, respectively. The measured high radionuclide content (^{226}Ra, ^{232}Th) in Bharati samples compared to Maitri samples is in conjunction with the reported high gamma dose rate in Bharati region (Bakshi et al. 2013). The measured values of uranium content in Maitri samples was observed to be four to five times higher than reported by Jojo et al. (1995). This could be due to the different sampling locations since the radium content of soils can show considerable spatial variability over a span of 20 years, both locally and regionally. These could in turn be due to difference in parent materials and in soil forming factors such as climate and weathering time. Soil development processes can lead to substantial redistribution of trace elements and radionuclides, such as radium, in the soil profile, thereby introducing variations in distribution with depth, as well as location (Meriwether et al. 1995; Van den Bygaart and Protz 1995).

The calculated values of gamma dose rate in Bharati Station were observed to vary from 128 to 264.1 nGy h^{-1} with average value of 198 ± 39 nGy h^{-1}. These calculated values of gamma dose rate are within the range as reported by Bakshi et al. (2013), though the average gamma dose rate measured using RadEye G gamma survey meter was about 300–400 nGy h^{-1} which is about two times higher than that of the calculated gamma dose rate in the present study. This increase in the measured gamma dose rate by Bakshi et al. (2013) is due to the combined contribution of both terrestrial gamma and cosmic rays, whereas the calculated gamma dose rate in the present study includes only terrestrial gamma. Compared to Bharati, average gamma dose rate in Maitri station was observed to be five times lower due to lower radionuclide content.

From Table 2, it is also observed that when the ^{226}Ra content is higher, the ^{232}Th content of the sample is also higher. Therefore, a correlation study was carried out between ^{226}Ra and ^{232}Th contents for the Bharati samples in Antarctica sand samples as shown in Fig. 7, which revealed a

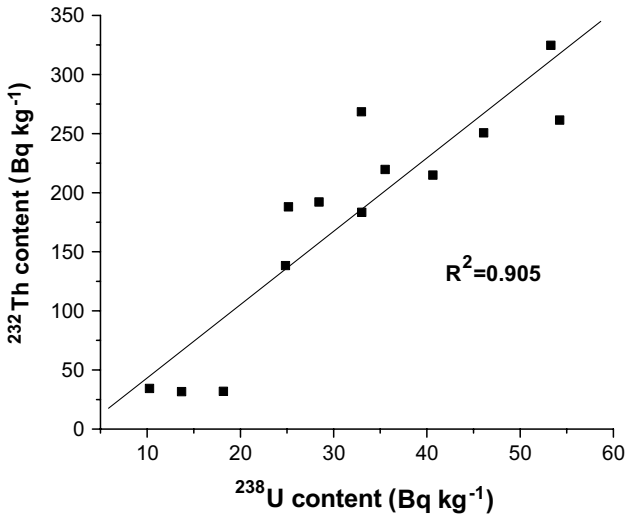

Fig. 7 Correlation between ^{232}Th content and ^{226}Ra content in Antarctica soil samples by gamma spectrometry

Table 2 Activity concentrations of ^{226}Ra,^{232}Th, ^{40}K, radium equivalent activity and dose rate in Antarctica sand samples

Sample code	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Radium equivalent activity (Bq kg^{-1})	Dose rate (nGy h^{-1})
B-1	54.2 ± 1.3	261.4 ± 2.4	1188.5 ± 7.9	519.5	236.9
B-2	24.8 ± 1.9	138.3 ± 4.5	735.3 ± 7.3	279.2	128.0
B-3	40.6 ± 2.8	214.9 ± 3.2	1019.2 ± 8.5	426.4	194.7
B-4	28.4 ± 1.9	192.1 ± 3.1	1002.2 ± 8.9	380.3	174.2
B-5	46.1 ± 1.8	250.6 ± 2.9	969.4 ± 7.2	479.1	217.3
B-6	35.5 ± 1.5	219.6 ± 2.4	1062.6 ± 6.5	431.3	197.1
B-7	25.2 ± 1.9	187.9 ± 3.2	961.3 ± 8.9	367.9	168.4
B-8	32.9 ± 2.0	268.4 ± 2.9	936.4 ± 7.1	488.8	220.9
B-9	33.0 ± 1.5	183.3 ± 2.3	1177.9 ± 7.0	385.8	178.2
B-10	53.3 ± 2.2	324.7 ± 3.7	907.57 ± 8.1	587.5	264.1
M-1	18.2 ± 1.6	31.8 ± 2.0	554.1 ± 9.7	106.4	51.3
M-2	10.3 ± 1.5	34.3 ± 2.0	478.3 ± 9.9	96.1	46.0
M-3	13.7 ± 1.5	31.6 ± 2.0	534.4 ± 6.9	100.0	48.2

B Bharati station, *M* Maitri station

positive correlation ($R^2 = 0.98$) similar to that observed in the radiometry of monazite-rich soil samples from HBRAs of Kerala (Derin et al. 2012). This strong correlation indicates that the sample population is dominated by ^{232}Th and ^{238}U bearing monazite minerals because in normal soils the correlation between Th and U is found to be weak (Kannan et al. 2002). A correlation study carried out between ^{232}Th and ^{40}K contents showed a negative correlation ($R^2 = -0.09$). This implies that ^{40}K content is not related to thorium-bearing minerals in the soil. Also, the ratio of activity concentrations(^{232}Th/^{226}Ra) for all soil samples of Bharati station varied from 4.8 to 8.1 with a mean (± SD) value of 6.1 ± 1.0. This trend is in agreement with that observed in the radiometry of soil samples from monazite-rich Kalpakkam beach sands with a range of 1.5–13.7 and mean of 5.5 (Kannan et al. 2002).

The radium equivalent activity computed using Eq. 9 was observed to vary from 279.2 to 587.5 Bq kg^{-1} in Bharati with an average value of 434.58 ± 87.88 Bq kg^{-1}, whereas in Maitri it was 100.8 ± 5.2 Bq kg^{-1}. These values are found to be lower than the criterion limit of 370 Bq kg^{-1} (OECD 1979) for the Maitri samples and slightly higher for the Bharati samples. Thus, does not pose much of any radiological hazard, considering the meager human habitation in Antarctica, compared to ~ 50 times higher values reported in samples from densely populated high background regions of Kerala in India (Derin et al. 2012).

A comparison of radionuclide content and dose rate obtained for Antarctica samples in the present study with the studies available in literature is shown in Table 3. It is observed that the ^{226}Ra content for Maitri samples was within the earlier reported range (Verma et al. 2006; Jojo et al. 1995) and for Bharati samples it was within the reported range for Normal Background Radiation Areas (NBRAs) of India and the World. But in case of ^{232}Th content, it was observed to be more than the reported range for Normal Background Radiation Areas (NBRAs) of India and the World (UNSCEAR 2000). The average dose rate at 1 m from the ground resulting from the radionuclide content is estimated to be 48.51 ± 2.65 nGy h^{-1} for Maitri station which is in agreement with that reported in other regions of Antarctica as well as world average value (Navas et al. 2005; Godoy et al. 1998). But for Bharati station, the average dose rate was estimated as 198.01 ± 38.77 nGy h^{-1} which is about three to four times higher than the global average value of 60 nGy h^{-1} (UNSCEAR 2000).

^{222}Rn/^{220}Rn progeny measurements

The measured ^{222}Rn/^{220}Rn progeny concentration at eight different indoor locations of Maitri station using DRPS and DTPS are shown in Table 4. The measured EETC was found to vary between 0.24 and 0.69 Bq m^{-3} with an average of 0.48 ± 0.14 Bq m^{-3} and the EERC was found to vary between 1.7 and 6.9 Bq m^{-3} with an average of 4.2 ± 1.9 Bq m^{-3}. The average values obtained for EETC and EERC in indoors are similar to that obtained in normal indoor conditions (Mayya et al. 2012). The EERC values are similar to that reported during the earlier expedition (Ramachandran and Balani 1995). Inhalation dose due to radon and thoron progeny calculated using Eq. 10 was found to be

Table 4 Measured EETC and EERC using DTPS and DRPS deployed at different indoor locations in Maitri station, Antarctica

Location no.	EETC (Bq m^{-3})	EERC (Bq m^{-3})	Inhalation dose (mSv)
1	0.46	3.1	0.133
2	0.40	1.7	0.090
3	0.53	4.4	0.175
4	0.58	7.0	0.248
5	0.69	4.2	0.188
6	0.41	6.9	0.226
7	0.24	2.9	0.103
8	0.52	3.1	0.140

Table 3 Comparison of results of radioactivity content in sand samples in the present study at the Indian stations with that of other regions of Antarctica, World average and Indian average

Area	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	J_M (mBq kg^{-1} h^{-1})	Absorbed dose rate in air (nGy h^{-1})	References
Antarctica (Bharati)	24.8–54.2	138.3–324.8	907.6–1189	3.44–7.86	128–264	Present study
Antarctica (Maitri)	10.3–18.2	31.59–34.33	478–554	2.41–3.15	46–51	Present study
Antarctica (Maitri)	2.47–6.17	3.28–12.3	–	–	–	Verma et al. (2006)
Antarctica (Maitri)	0.4–4.5	–	–	–	–	Jojo et al. (1995)
Antarctica Peninsula	5–36	4–31	125–810	–	–	Navas et al. (2005)
Antarctica (South Shetland Islands)	2.74–20.5	10–25	91–435	–	38–61	Godoy et al. (1998)
World	16–110	11–64	140–850	–	33–107	UNSCEAR (2000)
India	7–81	14–160	38–760	11–63	20–1100	UNSCEAR (2000)

0.163 ± 0.057 mSv for exposure period of 4 months. In comparison with the annual natural background dose of 1.3 mSv due to radon and thoron progeny, this value is quite low.

In case of outdoors, average EETC and EERC estimated using integrated sampler were obtained as 0.17 Bq m^{-3} and 2.2 Bq m^{-3}, respectively. From the measured EERC of 2.2 Bq m^{-3} and using the equilibrium factor of 0.6 for ^{222}Rn in outdoors (UNSCEAR 2000), the radon gas concentration is calculated to be around 3.7 Bq m^{-3} which is well within the mean global value of 5 Bq m^{-3} as reported in the lower troposphere (UNSCEAR 1993).

Conclusion

Baseline studies were carried out to explore the radiological characteristics of soil samples collected from two Indian Stations, Maitri and Bharati, at Antarctica based on the reported higher gamma exposure rates and soil radioactivity content. ^{222}Rn/^{220}Rn exhalation rates, radionuclide content, ^{222}Rn emanation coefficient and ^{222}Rn/^{220}Rn progeny concentration in indoor as well as outdoor locations were measured. The following conclusions can be drawn from this study.

1. The measured values of ^{222}Rn mass exhalation rate and ^{220}Rn surface exhalation rate of soil samples collected from Bharati and Maitri samples were observed to be within the normal reported range of exhalation rates of soil samples in literature. Good positive linear correlations were observed between ^{226}Ra content and ^{222}Rn exhalation rate, and between ^{232}Th content and ^{220}Rn exhalation rate.
2. Estimated ^{222}Rn emanation fraction of the sample grains ranged from 1.38 to 3.1% which is relatively lower compared to that obtained from other regions, which is attributed to the fact that the soils prevalent in these areas are mainly sands and loamy sands with larger grain size having lesser specific surface area (m^2/gm) and henceforth a lower radon emanation coefficient. Due to lower emanation coefficient, the overall exhalation rate of ^{222}Rn and ^{220}Rn was found to be within the normal range in spite of higher radioactivity content.
3. A positive correlation ($R^2 = 0.98$) observed between ^{226}Ra and ^{232}Th content and a negative correlation ($R^2 = -0.09$) observed between ^{232}Th and ^{40}K contents, a trend observed in radiometry of monazite rich beach sands in HBRAs of Kerala, indicate that the sample population is dominated by Th- and U-bearing minerals. Also, the ratio of activity concentrations (^{226}Ra/^{232}Th) was similar to that obtained in the radiometry of monazite-containing beach sand in HBRAs of Kerala. The radium equivalent activity values are found to be lower than the criterion limit of 370 Bq kg^{-1} for the Maitri samples and only slightly higher for the Bharati samples. Thus, does not pose much of any radiological hazard, considering the meager human habitation in Antarctica.
4. As a preliminary step towards measurement of ^{222}Rn and ^{220}Rn progeny concentration using indigenously developed DRPS/DTPS at the Maitri station, average EERC and EETC in indoors were found to be 4.2 Bq m^{-3} and 0.48 Bq m^{-3}, respectively, whereas, in outdoors EERC and EETC were found to be 2.2 Bq $m^{-3}$and 0.17 Bq m^{-3}. These values of EERC and EETC are within the global reported range. Inhalation dose due to ^{222}Rn and ^{220}Rn progeny was found to be 0.163 mSv which is quite low in comparison with the annual natural background dose of 1.3 mSv due to ^{222}Rn and ^{220}Rn progeny.

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