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A COMPARATIVE STUDY OF SOIL RADON AND THORON EXHALATION FROM FOUR VILLAGES OF KOLLAM, KERALA

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ABSTRACT

Exhalation rates of Radon and Thoron from surface soil were measured at four selected villages of Kollam district in Kerala, India, using a continuous radon monitor coupled to an accumulator system. Two villages (Chavara and Neendakara) in the coastal high background radiation area and the other two (Panmana and Ayanivelikulangara) were from interior normal background region. Environmental gamma dose, radon and thoron gas concentrations in the outdoors and exhalation of radon and thoron from surface soil were measured. Locations for measurement were selected on the basis of prevailing environmental gamma level. Measured rates of radon exhalation from surface soilin Chavarawere found to vary from 11Bq$m^{-2}h^{-1}$ to 301Bq$m^{-2}h^{-1}$ and those from Neendakara were from 17 Bq$m^{-2}h^{-1}$ to 317 Bq$m^{-2}h^{-1}$. Exhalation rates of Thoron were found to have a wide variation from 0.187 KBq$m^{-2}h^{-1}$to 26.6 KBq$m^{-2}h^{-1}$ in Chavara village and that in Neendakara village were found to vary from 0.218 KBq$m^{-2}h^{-1}$ to 144.3 KBq$m^{-2}h^{-1}$. Environmental gamma dose levelswere found vary from 0.17µSvh^{-1} to 4.75µSvh^{-1} in Chavara and from 0.31µSvh^{-1} to 7.63µSvh^{-1}in Neendakara. Radon exhalation rates from Panmana village varied from 12 Bq$m^{-2}h^{-1}$ to 151 Bq$m^{-2}h^{-1}$ and thoron exhalation rates varied from 0.35 KBq$m^{-2}h^{-1}$ to 22.76 KBq$m^{-2}h^{-1}$. Exhalation rates of radon in Ayanivelikulangara village varied from 56Bq$m^{-2}h^{-1}$to 189 Bq$m^{-2}h^{-1}$ and thoron exhalation rates varied from 6.57 KBq$m^{2}h^{-1}$ to 24.66 KBq$m^{-2}h^{-1}$. Gamma dose levelsin Panmana village were found to vary from 0.04 to 0.83 µSvh^{-1} and that in Ayanivelikulangara village was from 0.10µSvh^{-1} to 1.23µSvh^{-1}. The observations of exhalation ratesand gamma dose levels were found to have no correlation with each other. Radon exhalation rates from soil with the atmospheric radon (outdoor) were found to have a very weak correlation while for thoron, there were no correlation observed.

KEY WORDS *:* Radon, Thoron, Exhalation, Soil

INTRODUCTION

United nation's scientific agency - UNSCEAR – has already made it clear that the major contribution for natural radiation comes from Radon (UNSCEAR, 2000 & 2006). Inhalation of radon its progeny is the second largest reason for lung cancer (WHO, 2005 & 2009). Considerable difference in half-lives of radon, ^{222}Rn (3.85 days) and thoron,^{220}Rn (55.6s) not much studies have been conducted for thoron to conclude its role on the ill effects on human health. Having a very short decay period, thoron cannot travel far from the source. As compared with radon, thoron has progenies differing in half lives and energies of radiation. In this aspect the study of thoron along with radon is important in some regions like elevated background radiation areas like Kollam district of Kerala, India. This is the first time a detailed study of exhalation rates of radon thoron is conducted in this region.

In the present study, we carried out *in situ* exhalation rate measurements of the radioactive gases, radon and thoron from four villages in the coastal district of Kollam in Kerala state. Thoron and radon exhalation from ground is governed by on many factors such as radium activity levels in soil, soil porosity, soil temperature, soil water saturation, atmospheric pressure, geology of the area, surface wind and emanation coefficient (Hosoda *et al*., 2009). Owing to the short half-life period of thoron gas, the effective depth of source of thoron is much smaller than that of radon which emphasise the local variation in the rates of exhalation of thoron (UNSCEAR, 2000). In order to make measurements of rates of exhalation of radon and thoron levels, a detailedstudy wascarried outon the environmental gamma radiation dosesin the villages of Kollam district. Based on the data obtained,we selected four villages for the comparative study of the exhalation of the radioactive gases radon and thoron from soil. Out of the four villages, two villages namely Chavara and Neendakara has high background radiation while Panmana and Ayanivelikulangara are villages with normal background.

LOCALE

The four villages selected for the study were Neendakara (8.9443°N, 76.5402°E), Chavara (8.9908°N, 76.5404°E), Panmana (9.0078°N, 76.5412°E) and Ayanivelikulangara (9.0434°N, 76.5199°E).Of these, two villaages, Chavara and Neendakara are situated along the west southern coast of Kerala, which is awell-known high background radiation area(HBRA). The area is very rich inmonazite sand which comprises of orthophosphate of thorium and rare earths. Sand in the coastal region typically contains thorium oxide (~9 %) and uranium oxide (~0.35%) in addition to phosphorous pentoxide, rare earths, titanium oxide,

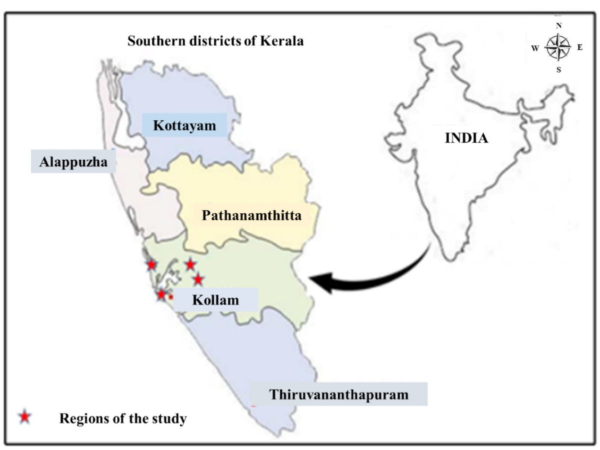

Fig. 1. Approximate locations selected for the radon and thoron exhalation studies in Kollam district

iron oxide,cerium oxide and silicon dioxide (Chougaonkar *et al*., 2004). The other villages arein theinterior areasin the vicinity of the coast. Therefore, the present investigation was made to examine exhalation rates from soil having different radioactive propertiesin a comparative manner. The regionhas two major industrial establishments. Kerala Minerals and Metals Limited (KMML) and Indian rare earths (IRE) are engaged in mining metals and minerals from the coastal sand.

Kerala Minerals and Metals produces high-grade titanium compounds such as titanium oxide pigment, hafnium ore, sillimanite and monazite. India Rare Earths working in the region is engaged in the mining and separation of heavy minerals of industrial importance such as limonite, rutile, zircon, sillimanite, garnet and monazite from beach sands. Monazite is source tem of thoron and radon. The residential houses in this region are very close to this coastal belt and hence thoron and radon source term analysis in this region is of relevance. Altogether 48 locations were selected for the exhalation rate measurement studies where gamma dose rate measurements. Environmental levels of radon and thoron gases were also determined using active methods.

MATERIALS AND METHODS

Assessment of gamma dose in the locations

Levels of atmospheric gamma absorbed dose rates were measured in all the sampling sites using a hand held gamma dosimeter (POLIMASTER, Model PM-1405). The PM-1405 can detect gamma rays in the energy range of 0.05–3 MeV and gamma dose in a wide range between 0.1 ìSvh^{-1} to 100 mSvh^{-1}. Gamma dose rates were determined using portable gamma dosimeter held at 1m from the ground. The locations for the exhalation rate study were selected based on the gamma dose meter readings.

Outdoor radon and thoron measurement

There are various active as well as passive techniques by which radon and thoron concentrations can be assessed. In present investigation, a scintillation based radon/thoron monitor (Smart Rn Duo) was employed. In this instrument, alpha particles emitted from radon/thoron inside the detector volume are detected by means of scintillation with ZnS(Ag). With the help of an algorithm based on radioactive growth and decay laws, the counts for each time interval are converted to radon/thoron concentration. The minimum detection limit of SMART RnDuo is 8 Bqm^{-3} at 1 sigma and has an upper detection limit of 50 $MBqm^{-3}$.

Exhalation rate measurements of radon and thoron

State of the art accumulator technique was employed for the assessment of exhalation rates of radon/thoron. Scientifically and technologically superior ZnS(Ag) based real time radon and thoron monitor, SMART Rn Duo, with a specially designed accumulator was used for the study. The device can automatically compensate the background counts resulting from the decay of radon/thoron progeny and thus able to quantity radon/thoron concentrations continuously. For the estimation of airborne radon air is sampled into the ZnS(Ag) scintillation cell through a "progeny filter" followed by a thoron barrier. Scintillations produced by the alpha particles originating from radon and its decay products inside the cell are continuously counted with the support of a photo multiplier tube and the allied counting electronics (Gaware *et al.*, 2011). The signals obtained are sorted out by a microprocessor unit as per the developed algorithm to display the concentration of radon.

The in-situ estimation of thoron exhalation from the surface beach sand was carried outemploying accumulator technique (Hosoda *et al*., 2009; Midhun *et al*., 2017). A sampler working in flow modewas linked to the pump inlet of the thoron monitor. Sampling pump was kept ON for initial 5 minutes to make an estimate of thoron and background. To ensure almost complete decay of thoron a delay of 5 minutes was given which was followed by last 5 minutes counting to measure of background counts for that cycle. Hence the intervention of long lived progeny of thoron (^{212}Pb, half-life of 10.6 hours) as well as the pulses due to radon (3.82 days) were not expected in the thoron monitor. Subtraction of background from gross counts for each cycle were taken care of. Direct scintillation with ZnS(Ag) does not have any interference from charge neutralizing species (such as humidity, CO_2, CH_4etc.). Therefore, the system does not require any drying. This strongly justifies the selection of this equipment for measurements in a location where humidity level is quite high. Relatively smaller accumulator was used for the estimation of thoron so as to confirm the assumption of uniform concentration in the accumulator. Thoron sampling from the accumulator was done'in flow mode' using a closed loop arrangement. The details of instrumentation and experimental procedure for the measurement and calculation of Radon and Thoron exhalation rates are discussed elsewhere (Midhun *et al*., 2017).

Accumulator was deployment with aburial depth of 1 cm in the surface soil. The measurement continued for 1hour at an interval of 15 minutes. Average of the last three readings was taken to assess the steady-state thoron concentration, C_e(Bq m^{-3}) in the accumulator. Obtained value of $C_e$was used to compute thoron flux, J_T ($Bqm^{-2}s^{-1}$) from the equation (1) obtained by the principle of mass balance (Sahoo and Mayya, 2010):

$$J_T = \frac{C_e V \lambda}{A} \quad .. (1)$$

where, λ is the decay constant (0.0126 s^{-1}) forthoron, Vis the total volume comprising of accumulator, detector and tubing volume(m^3) and Ais the surface area (m^2) of the surface sand matrix sealed off by the accumulator.

The estimation of in-situ radon rates exhalation was done by accumulator methodby applying analysis of exponential growth curve (Sahoo and Mayya, 2010). A cylindrical accumulator of 15 cm diameter and 15 cm height was coupled with the continuous radon monitor. The accumulator was buried firmly on the surface soil to a depth of 1 cm. The measuring device was used in diffusion mode for radon measurement. The mixed gas entry point to the instrument chamber was at a height of 15 cm from the soil surface. This was done to get rid of thoron entry (if present in soil) in to the accumulator. Radon gas concentration builds up inside the accumulator was recorded in every 15 minutes and the measurements continued for 5 – 7 hours. The radon concentrations (Bqm^{-3}) estimated with time elapsed (t) with respect to start of the accumulation was plotted. The equation for radon growth in a typical accumulator is given by the exponential relation (Sahoo *et al*., 2014):

$$C(t) = \frac{J_R}{V\lambda_e}\left(1 - e^{-\lambda_e t}\right) + C_0 e^{-\lambda_e t} \quad .. (2)$$

Here, the terms C(t) is the concentration of radon in the accumulator (Bq m^{-3}) at time t,$J_R$is the estimated radon flux from soil (Bq m^{-2} h^{-1}), V is volume of the accumulator (m^3),$ë_e$is the effective decay constant (h^{-1}) of radon. The effective decay constant is the sum of radon decay constant, leakage rates (if any) and back diffusion rate for the set up.

A denotes the area of soil surface covered by accumulator (m^2) and C_o represents the radon concentration inside the accumulator at initial condition, t=0. Experimentally obtained radon build up data can be plotted against the exponential decay equation:

$$Y(x)=Y_o + A_1 e^{-x/t1} \quad .. (3)$$

By plotting the graph we can obtain the fitting parameters Y_o, A_1 and t_1. Using the equations (2) and(3) we get the radon surface exhalation rate, $J_R=Y_0V\lambda_e/A$ and effective decay constant $\lambda_e = 1/t_1$. Figure. 2 shows the typical plot of radon build up inside the accumulator.

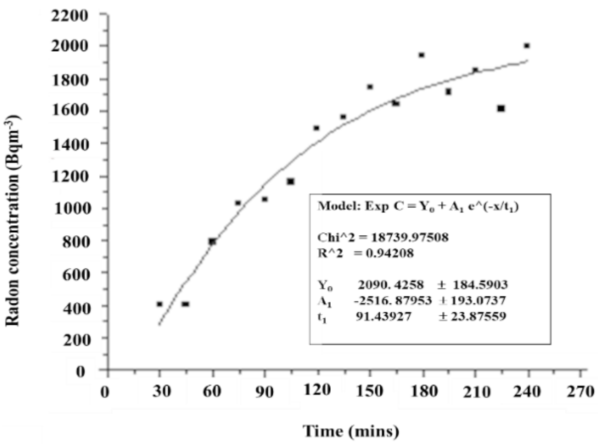

Fig. 2. Build-up of radon in the accumulator chamber

In each region, twelve locations were demarcated to conduct the radon and thoron exhalation measurements. While making measurements in each location environmental gamma dose measurements were also taken with the dosimeter.

RESULTS AND DISCUSSION

Three types of measurements – environmental gamma dose, radon and thoron gas levels and exhalation of radon and thoron from surface soil - were made in four villages in twelve locations each. Summary of the investigations in the four villages are presented in the Table 1. Details with regard to the concentrations of outdoor radon and thoron, gamma dose rates in the environment, minimum, maximum and average values of measured radon and thoron exhalation rates of radon and thoron in Chavara, Neendakara, Panmana and Ayanivelikulangara villages are presented.

The data indicate that the measured values have a wide range indicating the vast heterogeneity of soil radioactivity in the region. Therefore, it is meaningless to figure out a representative value for the measured quantities. However, an attempt was made to look for any correlation between the measured quantities.

Figure 3 is a graphical representation of the variation of radon and thoron exhalation rates in the four villages. The gamma dose levelswere found to vary from 0.17μSvh^{-1} to 4.75μSvh^{-1} in Chavara and 0.31μSvh^{-1} to 7.63μSvh^{-1} in Neendakara. The graph shows the exhalation rates of thoron are greater than that of radon exhalation which emphasise the

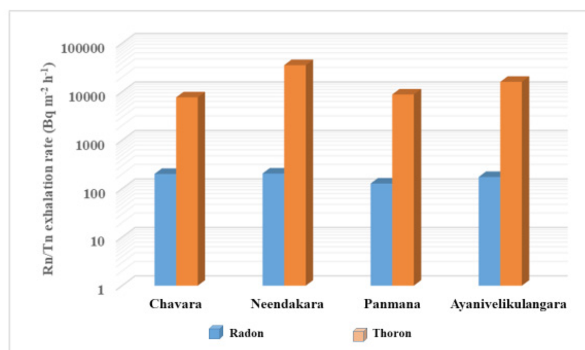

Fig. 3. Histograms for the average rates of radon and thoron exhalation in the four villages

Table 1. Average, maximum and minimum of measured exhalation rates of radon and thoron in four villages of Kollam district, Kerala.

Name of locations	Range of outdoor Radon Conc. Bq m^{-3}	Range of outdoor Thoron Conc. Bq m^{-3}	Gamma dose range* μSvh^{-1}	Radon exhalation rate (Bqm^{-2}h^{-1})			Thoron exhalation rate (Bqm^{-2}h^{-1})		
				Min.	Max.	Ar. mean	Min.	Max.	Ar. mean
Chavara	24 to 89	36 to 112	0.17to 4.75	11	301	202	187	26661	7670
Neendakara	32 to 94	44 to 129	0.31 to 7.63	17	317	206	218	144267	35429
Panmana	8 to 34	8 to 24	0.04 to 0.83	12	151	128	345	22762	8788
Ayanivelikulangara	8 to 46	10 to 36	0.10 to 1.23	56	189	175	6575	24656	16046

* As measured by the field dosimeter PM140

presence of thorium deposits in the beaches. An attempt was made to find correlation, if any, between the environmental gammas dose rates with the exhalation rates of radon from soil. It can be seen from the Figure 3 that there is no noticeable correlation between radon exhalation and gamma dose rates. Such a study would be meaningless for thorn since the it has a very short half-life.

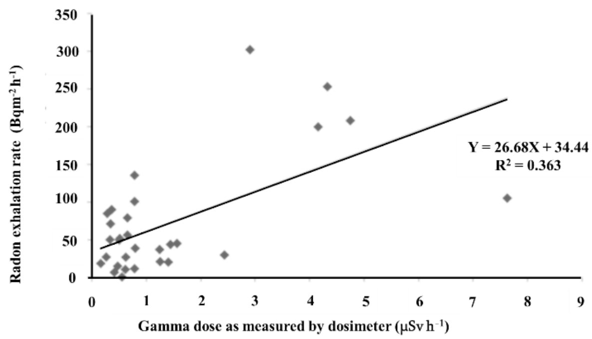

Fig. 3. Scatter diagram of radon exhalation rate against gamma dose in beach regions (Chavara and Neendakara)

It was noticed that the estimated rates of exhalation of radon and thoron were minimum where the area in which the dredging was done by KMML or IRE for the mineral extraction from soil. Radon exhalation rate was found maximum at the location Kovilthottam in Neendakara village. Moisture content of soil can also influence thoron and radon exhalation marginally (Hosoda *et al.*, 2007). The exhalation rate of thorn and radon were found to be influenced by the physical properties of soil. The average values of radon exhalation rates as well as thoron exhalation rates at all the villages were found greater than the reported global average value of 94.32 for radon exhalation and 3600 Bqm^{-2}h^{-1} for thoron exhalation (UNSCEAR, 200; Ahmad *et al.*, 2014).

Relationship between atmospheric (outdoor) radon and radon exhalation rates from soil in the forty-eight locations were studies for any possible correlation. It was found that there exist only weak correlations in all the four villages between atmospheric radon and exhalation rates of radon from soil. This could be due many confounding factors which influences the both radon exhalation from soil and more likely the ambient environmental conditions which disperses the released gas to the atmosphere.

CONCLUSION

The exhalation rates of thoron were greater than that the radon exhalation rates in the four villages. This could be due to high thorium levels present the surface soil in the region. Both exhalation rates show a poor correlation with the radioactivity content and gamma levels. Outdoor radon and thoron concentrations are considerably higher in the coastal villages of Chavara and Neendakara as compared with the interior villages of Panmana and Ayanivelikulangara. Environmental gamma dose rates are independent of radon exhalation rates from

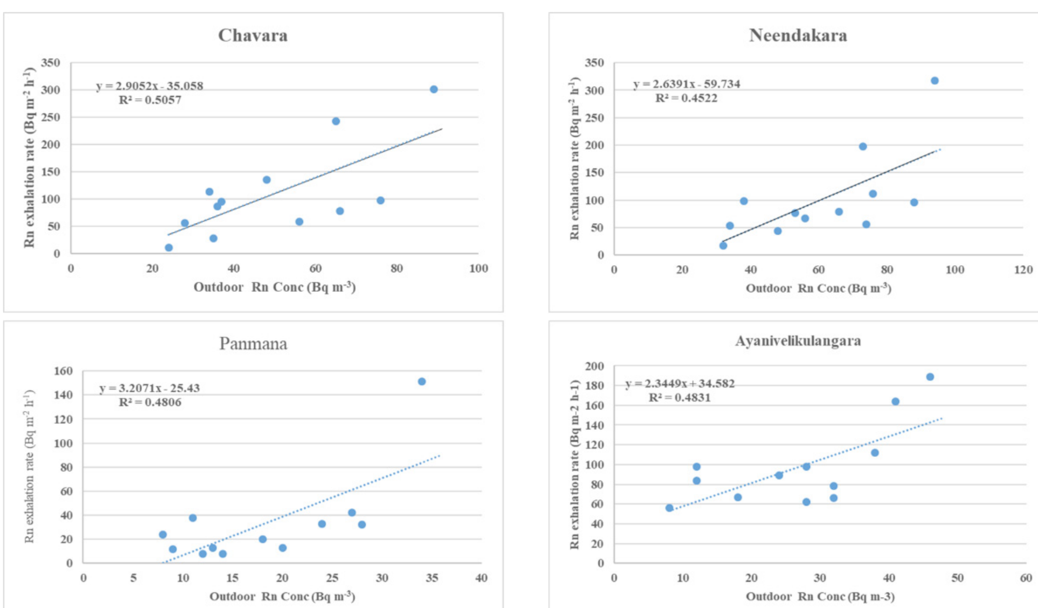

Fig. 4. Relationship of radon exhalation rates from soil with outdoor radon concentration in the four villages

soil. There exists a weak correlation between the environmental radon concentration (outdoor) and radon exhalation rate from soil. The average values of radon exhalation rates as well as thoron exhalation rates at all the villages were found greater than the reported global average.

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