Study of radon exhalation from soil to ascertain the elevated levels of background radiation of Bangladesh

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Abstract. The outdoor environmental background radiation level in normal area of Bangladesh ranged between 110 and 445 nGy.h^{-1} was observed in a countrywide background radiation survey using β-γ radiation survey meter. The United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) reported to the united Nations General Assembly in its 1988 report that the outdoor country averaged environmental background radiation level in air in 23 countries representing about one half of the world population were found to range between 24 and 85 nGy.h^{-1}, with a mean value of 55 nGy.h^{-1}, which is one fourth of the radiation dose level of Bangladesh. In the present study, activity concentration of radium and radon radionuclides was measured in the soil samples of the country and measured their radon emanation coefficient, and radon mass exhalation and radon area exhalation rates with an aim to evaluate the cause of high background radiation level in Bangladesh. The result shows that the concentration of ^{226}Ra in most of the soil samples was higher than that of world average value, and the concentration of ^{224}Ra, a daughter of ^{232}Th-series, is more than two times higher than the world average value. The activity of ^{40}K was found higher than both the range and average world value in most of the samples. The results of ^{222}Rn emanation coefficients, ^{222}Rn mass and area exhalation rates are very significant. Values of radon emanation coefficients for the soil samples in a number of locations were found in excess of world average value. The ^{222}Rn emanation coefficients was found in the range 3.10 – 44.68%, the mass exhalation rates was found in the range 2.50 – 27.84 μBq.kg^{-1}.sec^{-1}, and the area exhalation rates was found in the range 3.15 – 38.15 mBq.m^{-2}.sec^{-1}. Due to high concentrations of radium and potassium in soil and the high radon exhalation rate the external and internal doses are also higher. The total external dose from soil was found higher than the reported results of most of the countries of the World. This result would be helpful to assess the radiation exposure to human, animal and plants, and radiation related cancer risk.

1. INTRODUCTION

The assessment of the radiation doses in human from environmental radiation is of special importance because radiation is harmful to man and environment. The major contribution to the various radiation exposures received by mankind comes from natural sources. Natural irradiation has been exposing the whole population of the world at a relatively constant rate over a long period of time. Large-scale surveys have been carried out in 23 countries in order to estimate average nation-wide exposures to outdoor external gamma-radiation, representing about one half of the world population, showed that country-averaged outdoor-absorbed dose rates in air were found to range between 24 and 85 nGyh^{-1}, with an arithmetic mean of 55 nGyh^{-1} [1]. Inhalation of short-lived decay products of radon (^{222}Rn) accounts on average for one half of the effective dose equivalent from all natural sources of radiation and may sometimes lead to doses high enough to cause concern for human health [2]. The sources of natural radiations are the members of three well-known natural radioactive series namely thorium, uranium and actinium series associated with their progeny [1]. Radon isotopes are the members of the natural radioactive series. Radon isotopes diffuse from the earth into the atmosphere producing a number of short-lived decay products. The concentration of the two radon isotopes in air is highly variable, and determined largely by the concentration of their precursors ^{226}Ra and ^{232}Th in the soil. Atmospheric concentrations are expectedly greatest over areas in which the soil is rich in radium and thorium. Diffusive ^{222}Rn migration in porous media normally does not exceed several meters, while ^{220}Rn only reaches some centimeters. The diffusive ^{222}Rn component in soil gas can be calculated from the specific ^{226}Ra activity, specific density, effective porosity and radon escape/production ratio of soils and rocks [3]. In Bangladesh the external background radiation levels lie between 1.0 and 3.9 mSv.y^{-1} with an average value of 2 mSv.y^{-1} excluding Cox's Bazar beach sand areas where an average value of 13 mSv.y^{-1} with a range of 2.6-44.0 mSv.y^{-1} was observed [4,5]. In the present study, activity concentration of radium and radon radionuclides was measured in the soil samples of the country and measured their radon emanation coefficient, and radon mass exhalation and radon area exhalation rates with an aim to evaluate the cause of high background radiation level in Bangladesh.

2. METHODS AND MATERIALS

In the present study, 150 samples of soil were collected from upper layer of soil up to 15 cm depth by using soil corer from the several districts of Bangladesh. The samples were cleaned, dried in air and crushed to fine powder. The powdered soils were filled into cylindrical plastic containers (dia. 6.5 cm, ht. 7.5 cm) of active volume 150 ml. To determine the fraction of ^{222}Rn could escape from the material, activity measurements were made on ventilated samples before they were baked and before the cans were sealed. After the measurements, the samples were then baked and sealed for determination of radionuclide contents. The difference between the total amount of ^{222}Rn generated by the material (^{226}Ra activity of the 4-week sealed samples) and the ^{222}Rn trapped in the material (^{214}Pb and ^{214}Bi activity of the ventilated samples) yielded the amount of ^{222}Rn that has escaped [6]. The fraction of radon produced, which can escape from the solid matrix of soil, is the emanation coefficient of soil [1]. Only this fraction of ^{222}Rn can diffuse through the soil and escape to the open air. The emanation rate is the product of the emanation coefficient and ^{222}Rn-production rate. The containers were sealed tightly and wrapped with thick vinyl tapes around their screw necks. The samples were stored for four weeks to allow ^{226}Ra and ^{232}Th to reach in secular equilibrium with their respective daughters [7], then the samples were counted using a p-type coaxial high purity germanium (HPGe) detector of active volume 132 cc, relative efficiency 35% and resolution 1.8 keV (FWHM) at 1.33 MeV γ-energy of ^{60}Co coupled with PC based multichannel analyser (PCMCA). The γ-ray energy regions selected for the corresponding radionuclides were 583 and 2614 keV of ^{208}Tl, and 911 and 969 keV of ^{228}Ac for the determination of ^{224}Ra; 295 and 352 keV of ^{214}Pb and 609.3, 1120 and 1764.5 keV of ^{214}Bi for the determination of ^{226}Ra, and 1460 keV for ^{40}K [7,8]. Measurements were carried out in a low background lead shielding arrangement. The efficiencies of the detector for the corresponding gamma energies were measured using IAEA reference samples RGU-1, RGTh-1 and RGK-1 of uranium ore, thorium ore mixed in silica matrix and potassium sulfate respectively, at identical sample counting geometry [9]. The concentrations of corresponding radionuclides were calculated from the net peak area.

3. RESULTS AND DISCUSSION

The mean results of activity concentrations of ^{226}Ra, ^{224}Ra and ^{40}K radionuclides in soil and derived radiation hazard parameters, the radon emanation coefficients and radon mass and area exhalation rates of soils are shown in Table 1. In the soil, the activity of ^{226}Ra ranged from 7.80 – 66.52 Bq.kg^{-1}, ^{224}Ra ranged from 27.0 – 98.95 Bq.kg^{-1} and the ^{40}K ranged from 218 – 1453 Bq.kg^{-1}. In most of the samples values of ^{226}Ra found are higher than the world average value of 25 Bq.kg^{-1} [1], and the values of ^{224}Ra, a daughter product of ^{232}Th-series, is more than two times higher than the world average value. The ^{40}K, a significant dose contributor to the environment and an naturally abundant primordial radionuclide, was found in high contents than the range and mean of world value in most of the locations. The world range and average ^{222}Rn emanation coefficients are 2-80% and 20% respectively [1]. In the present work, ^{222}Rn emanating coefficient value in a number location was found in excess of world average value. The ^{222}Rn emanation coefficient ranged from 3.10 – 44.68% and ^{220}Rn emanation coefficient ranged from 0.13 – 18.48%. The mass activity emanation rate, expressed in Bq kg^{-1}s^{-1} and defined as [1]

$$R_m = \lambda_{Rn} C_{soil,Ra} F_r$$

where R_m is the mass exhalation rate in Bq.kg^{-1}.sec^{-1}, λ_{Rn} is the decay constant of radon, C_{Rn} is the concentration of Ra in soil and F_r is the emanation coefficient. The ^{222}Rn mass exhalation rate is ranged from 2.50 – 27.84 μBq.kg^{-1}.sec^{-1} and the ^{220}Rn mass exhalation rate is ranged from 1.02 – 17.17 mBq.kg^{-1}.sec^{-1}

The area exhalation rate, defined as the activity transfer rate per unit area at the soil-air interface, is expressed as [1]:

$$R = \lambda_{Rn} F_r C_{soil,Ra} \rho_{soil} L_{Rn}$$

where R is the area exhalation rate in Bq m^{-2} s^{-1}; λ_{Rn} is the decay constant of ^{222}Rn (2.1×10^{-6} s^{-1}); F_r is the emanating power; $C_{soil,Ra}$ is the activity mass concentration of ^{226}Rn in soil (Bq kg^{-1}); ρ_{soil} is the soil density (kg m^{-3}); and L_{Rn} is the diffusion length of radon in soil (m). The ^{222}Rn area exhalation rate is ranged from 3.15 – 38.15 $mBq.m^{-2}.s^{-1}$ and ^{220}Rn area exhalation rate is ranged from 0.30 – 11.71 $mBq.m^{-2}.s^{-1}$. The world average value of ^{222}Rn area exhalation rate is 16 $mBq.m^{-2}.sec^{-1}$. Area exhalation rate in a number of soil samples were found higher than that of world average value.

The external dose rate at 1 m above the ground surface due to the natural radionuclides in soil is ranged from 46 – 137 $nGy.h^{-1}$ with an average dose of 82 $nGy.h^{-1}$ were observed. The total external dose from soil was found higher than the reported results of most of the countries of the World. In India, the absorbed dose rate in air from all the natural sources is 20 - 1100 $nGy.h^{-1}$ and its average is 55 $nGy.h^{-1}$. In China, the concentration of ^{226}Ra was reported by Ziqianj *et al* is 18.2 - 135 $Bq.kg^{-1}$ and the dose rate in air 6.07 - 15.1 $nGy.y^{-1}$. The reported highest absorbed dose rate area of the World are Norway and Sweden, there ranges are 20 - 1200 and 18 - 4000 $nGy.y^{-1}$. The range of absorbed doses of Austria is 20 - 150, France is 10 - 250, Germany 4 - 350, Italy 7 - 500 and Romania is 32 - 210 $nGy.h^{-1}$ [1]. Due to high concentrations of radium and potassium in soil and high exhalation rate of radon the external and internal doses are also higher. This result would be helpful to assess the radiation exposure to human, animal and plants, and radiation related cancer risk.

Table 1. The mean activity concentrations of ^{226}Ra, ^{224}Ra and ^{40}K radionuclides in soil of several districts of Bangladesh and derived radiation hazard parameters.

Sl.	Location	# Samples	^{226}Ra, $Bq.kg^{-1}$	^{224}Ra, $Bq.kg^{-1}$	^{40}K,$Bq.kg^{-1}$
1	Chittagong	36	22.8 ± 6.1	44.8 ± 17.5	766 ± 279
2	Cox's Bazar	9	15.9 ± 8.9	36.5 ± 6.3	581 ± 62
3	Moheshkhali	9	20.9 ± 3.4	30.5 ± 2.5	457 ± 10
4	Comilla	9	40.5 ± 1.3	71.8 ± 5.3	741 ± 131
5	Sylhet	15	23.2 ± 4.1	61.2 ± 25.3	392 ± 131
6	Jaipurhat	12	52.6 ± 8.2	82.6 ± 8.6	836 ± 242
7	Sirajganj	15	48.6 ± 7.0	72.6 ± 18.2	904 ± 115
8	Pabna	15	35.4 ± 2.7	66.8 ± 14.7	761 ± 182
9	Bogra	15	39.2 ± 16.4	62.8 ± 16.6	689 ± 203
10	Rajshahi	15	31.5 ± 4.4	64.0 ± 14.5	468 ± 241
		Mean	33.1 ± 12.3	59.4 ± 16.8	660 ± 175

Table 1. (Contd...)

Sl.	^{222}Rn Emanation coefficient Fr (%)	^{220}Rn Emanation coefficient Fr (%)	^{222}Rn Mass exhalation rate, μBq. kg^{-1}.s^{-1}	^{220}Rn Mass exhalation rate, mBq. kg^{-1}.s^{-1}	^{222}Rn Area exhalation rate, mBq. m^{-2}.s^{-1}	^{220}Rn Area exhalation rate, mBq. m^{-2}.s^{-1}
1	13.4 ± 6.4	1.09 ± 0.9	6.3 ± 1.4	6.0 ± 1.2	8.5 ± 4.6	1.7 ± 3.1
2	16.0 ± 3.4	1.06 ± 0.5	6.8 ± 1.3	4.7 ± 1.5	9.2 ± 1.7	6.3 ± 2.0
3	18.5 ± 1.8	0.93 ± 0.2	8.2 ± 2.0	3.6 ± 0.9	10.9 ± 2.7	4.7 ± 1.2
4	6.2 ± 0.8	0.36 ± 0.2	5.2 ± 0.5	3.2 ± 1.3	7.0 ± 0.7	3.4 ± 2.8
5	17.9 ± 5.5	6.87 ± 1.6	9.4 ± 1.8	4.5 ± 12	12.5 ± 3.2	5.8 ± 3.2
6	13.5 ± 2.9	0.5 ± 0.09	13.3 ±2.2	4.7 ± 0.5	18.2 ± 2.4	1.4 ± 0.2
7	16.3 ± 2.8	0.5 ± 0.2	17.5 ±6.7	4.5 ± 0.8	23.5 ± 3.0	1.3 ± 0.2
8	5.8 ± 5.0	1.4 ± 0.8	6.6 ± 3.0	11.9 ± 2.2	8.8 ± 4.5	3.4 ± 1.7
9	8.9 ± 2.9	0.6 ± 0.1	6.58 ±3.0	4.8 ± 1.2	8.8 ± 4.5	4.5 ± 1.0
10	17.9 ± 2.8	0.3 ± 0.07	11.7 ±0.6	2.5 ± 0.7	15.7 ± 0.7	3.4 ± 1.0
Mean	13.5 ± 4.8	1.36 ± 1.9	9.2 ± 3.9	5.0 ± 2.6	12.3 ± 5.3	3.6 ± 1.8

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