

Occurrences of NORMS and ^{137}Cs in soils of the Singhbhum region of Eastern India and associated Radiation Hazard

A. CHAKRABARTY¹, R.M. TRIPATHI¹, V.D. PURANIK¹

(Manuscript received 11 July 2008, accepted 17 September 2008)

ABSTRACT Naturally occurring radioactive materials (NORMS) and fallout ^{137}Cs were estimated in surface soils from the highly mineralized Singhbhum region of Eastern India. The activity concentrations varied from 50.67-109.14 Bq/kg (^{238}U), 48.12-142.55 Bq/kg (^{226}Ra), 28.73-89.78 Bq/kg (^{232}Th), 494.84-1121.36 Bq/kg (^{40}K) and 1.8-7.48 Bq/kg (^{137}Cs). Correlation between radionuclides was studied. $^{232}\text{Th}/^{238}\text{U}$ concentration (Clark value) was also calculated. Radiation hazard for the samples was assessed by radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}) and absorbed gamma dose rate (D). Ra_{eq} values ranged from 216-263 Bq/kg, H_{ex} values from 0.58-0.71 Bq/kg, H_{in} values from 0.73-1 Bq/kg and D values from 100-121 nGy/h. Calculated and observed dose rates in air were compared with the help of parametric and non-parametric tests. The tests showed significant difference between the calculated and observed dose rates.

Keywords: natural radioactivity / activity concentration / radiation hazard / dose rate

RÉSUMÉ La présence de MRON et de ^{137}Cs dans les sols de la région de Singhbhum, en Inde de l'est. Le risque associé à leur rayonnement.

Nous avons évalué les matériaux radioactifs d'origine naturelle (MRON) et le ^{137}Cs des retombées, dans les sols de surface venant de la région hautement minéralisée de Singhbhum, en Inde de l'est. Les concentrations de l'activité variaient de (50,67 à 109,14) Bq/kg (^{238}U), (48,12 à 142,55) Bq/kg (^{226}Ra), (28,73 à 89,78) Bq/kg (^{232}Th), (494,84 à 1121,36) Bq/kg (^{40}K), et (1,8 à 7,48) Bq/kg (^{137}Cs). Nous avons étudié la corrélation entre les radionucléides. La concentration ($^{232}\text{Th}/^{238}\text{U}$), ou valeur de Clark, a été également calculée. Le risque radiatif venant des échantillons a été évalué par l'activité radium équivalente (Ra_{eq}), par l'index de risque externe (H_{ex}), par l'index de risque interne (H_{in}) et par le débit de dose gamma absorbé (D). Les valeurs de Ra_{eq} allaient de 216 à 263 Bq/kg, les valeurs de H_{ex} de 0,58 à 0,71 Bq/kg, les valeurs de H_{in} de 0,73 à 1 Bq/kg, et les valeurs de D de 100 à 121 nGy/h. Nous avons comparé les débits de dose dans l'air calculés et observés, en nous aidant d'essais paramétriques et non-paramétriques. Les essais ont montré qu'il existe une différence significative entre les débits de dose calculés et observés.

¹ Environmental Assessment Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, India.

1. Introduction

The external radiation exposure arises mainly due to cosmic rays and from terrestrial radionuclides. Terrestrial radiation, mainly originates from the primordial radionuclides formed in the early stage of the formation of the solar system (Al-Jundi, 2002). Gamma radiation arising from naturally occurring radioactive materials (NORMS), such as ^{40}K , ^{238}U , ^{235}U and ^{232}Th series and their decay products, existing at trace levels in all ground formations, is the principal external source of radiation to the human body (Karahana *et al.*, 2000; Tzortzis *et al.*, 2004; UNSCEAR, 1993). Uranium has two primary isotopes ^{238}U (half-life: 4.5×10^9 years) and ^{235}U (half-life: 7.04×10^8 years) which occur in the proportion 99.27% (^{238}U) and 0.72% (^{235}U), respectively. Both exhibit long and complex decay series. ^{232}Th (half-life: 1.4×10^{10} years) is another naturally occurring radionuclide with a complex decay chain. Potassium has three isotopes (^{39}K , ^{40}K and ^{41}K), ^{40}K (half-life: 1.28×10^9 years) being the only radioactive isotope having an abundance of 0.012% of total potassium.

Absorbed dose rate in air, at sea level, from cosmic radiation is about 30 nGy/h for the Southern hemisphere (UNSCEAR, 2000). The levels of terrestrial environmental radiation in an area are related to the geological condition of that area and also to the content of Th, U and K in the rocks from which the soils originate (Tzortzis *et al.*, 2004). Hence, the levels of natural environmental radioactivity and the associated external exposure due to gamma radiation are observed to be at different levels in the soils of different regions in the world (Florou *et al.*, 1992; UNSCEAR, 2000). Thus, concentrations of elements present in soils on top of sedimentary materials are diverse; depending on their concentration in the eroded substance and also on the physico-chemical phenomena associated with sedimentation (Martinez-Aguirre *et al.*, 1997). Considering natural radioactivity, igneous rocks of granitic composition are strongly enriched in Th and U (an average of 15 ppm Th and 5 ppm U), compared to rocks of basaltic or ultramafic composition (<1 ppm of U) (Faure, 1986; Ménager *et al.*, 1993). Igneous rocks show higher radiation levels compared to sedimentary rocks. But there are exceptions as well, like; some shales and phosphate rocks contain relatively higher content of radionuclides (UNSCEAR, 2000).

Almost 95% of the world's population is assumed to live in areas of normal background radiation, where outdoor exposures range from 24 to 160 nGy/h (UNSCEAR, 1993). As external radiation exposures from naturally occurring radionuclides contribute nearly 25% of the average annual dose to the human body from all radiation sources, considerable attention has been given to low-level exposures arising due to naturally occurring radionuclides, particularly ^{238}U , ^{232}Th and ^{40}K (Higgy, 2002).

The Singhbhum thrust belt of Eastern India is the area under investigation. This area is known for hosting vein/disseminated type natural uranium deposits (Dikshitulu *et al.*, 1997) and is characterized by higher concentration of NORMS. Mining of low grade uranium, iron and copper minerals has been carried out at different locations in this region. Here we present studies on distribution of NORMS and fallout ^{137}Cs in the soils of this region and the associated radiological hazard.

2. Materials and methods

2.1. Sample collection and preparation

Surface soil samples were collected from the Singhbhum thrust belt of Eastern India. Multiple samples were collected from 0-5 cm depth at a location and homogenised to make one representative sample. Samples were dried at 105 °C, after removing vegetation, pebbles and stones. Samples were ground, homogenised and sieved to <2000 μm size. Weighed samples (approximately 300 g) were sealed in polyethylene bottles of 6.5 cm diameter and 7.5 cm height. Samples were sealed for one month so that secular equilibrium is attained between the daughter nuclides of ^{222}Rn and ^{220}Rn .

2.2. Instrumentation and calibration

A high-purity vertical Germanium detector was used for all the measurements. The system consisted of an n-type HPGe detector having 50% relative efficiency with respect to 7.6 cm \times 7.6 cm NaI(Tl) detector at 1332 keV of ^{60}Co gamma energy measured at 25 cm and associated electronics coupled with 8K MCA. Spectrum analysis was done by PHAST software (Electronics Division, BARC). IAEA Certified Reference Materials (CRMs), RGU-I and RGTh-I, were used for the energy and efficiency calibration of the High Resolution Gamma Spectrometric System (HRGs).

2.3. Natural radioactivity measurement

Samples were counted in polyethylene bottles of the same geometry as the CRMs. The γ -transitions observed for radionuclide determination are given in Table I. To determine the contribution of ambient background, an empty container, of the same dimensions used for the samples and with the same geometrical conditions, was counted on the detector. Sample and background spectrum were all acquired for 60,000 seconds.

TABLE I
Gamma energies used for analysis of radionuclides.
Énergies gamma utilisées pour analyser les radionucléides.

Radionuclide	Measured nuclide	Energy (keV)	Branching intensity (%)
^{238}U	^{234}Th	63.29	4.821
^{226}Ra	^{226}Ra	186.2	3.59
^{232}Th	^{208}Tl	583.19	30.369
^{232}Th	^{208}Tl	2614.53	35.64
^{232}Th	^{228}Ac	911.2	25.8
^{232}Th	^{212}Pb	238.63	43.3
^{40}K	^{40}K	1460	10.7
^{137}Cs	^{137}Cs	661.6	85.1

TABLE II
Activity concentrations (Bq/kg) of radionuclides in soils.
Concentrations de l'activité (Bq/kg) de radionucléides dans les sols.

Sample code	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
1	103.80 ± 12.66	131.36 ± 3.28	28.73 ± 1.19	671.32 ± 6.71	2.74 ± 0.23
2	62.98 ± 5.48	79.69 ± 3.35	68.12 ± 2.80	685.76 ± 9.60	6.58 ± 0.43
3	70.82 ± 3.75	78.52 ± 2.75	89.78 ± 2.03	539.66 ± 5.94	5.44 ± 0.33
4	52.60 ± 5.26	54.86 ± 2.19	76.90 ± 1.82	670.71 ± 6.04	3.65 ± 0.19
5	109.14 ± 10.04	142.55 ± 2.7	57.35 ± 1.34	494.84 ± 4.45	1.80 ± 0.15
6	50.67 ± 6.79	48.12 ± 4.57	68.76 ± 1.38	1121.36 ± 6.73	7.48 ± 0.33

3. Results and discussion

^{238}U , ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs activity concentrations, in Bq/kg, in the soil samples are given in Table II. The activity concentrations varied from 50.67-109.14 Bq/kg (^{238}U), 48.12-142.55 Bq/kg (^{226}Ra), 28.73-89.78 Bq/kg (^{232}Th), 494.84-1121.36 Bq/kg (^{40}K) and 1.8-7.48 Bq/kg (^{137}Cs).

Radionuclide activities obtained from work conducted worldwide have been tabulated in Table III. The data reported for Orissa (Mohanty *et al.*, 2004) are for a high background radiation area. The obtained results in the current study are higher than the worldwide median activity concentrations (UNSCEAR, 2000) of 35, 30 and 400 Bq/kg for ^{238}U , ^{232}Th and ^{40}K respectively except ^{232}Th activity concentration of 28.73 Bq/kg in sample 1. The presence of naturally occurring vein/disseminated type uranium mineralization in this region is the main factor for marginally elevated levels of natural radioactivity in the soils. In Table IV activity

TABLE III
Reported values of gamma activity (activity Bq/kg), from work conducted worldwide.
Valeurs publiées d'activité gamma (activité en Bq/kg) dans des recherches effectuées dans le monde entier.

Region	^{238}U	^{232}Th	^{40}K	Reference
Rajasthan, India	30-78	43-106	50-137	Nageswara <i>et al.</i> (1996)
Amman, Jordan	56.4	28.8	501.3	Ahmad <i>et al.</i> (1997)
Karak, Jordan	228.9	27.2	410.2	Ahmad <i>et al.</i> (1997)
Istanbul, Turkey	21	37	342	Karahan <i>et al.</i> (2000)
Costal area, Aegean sea, Greece	93 ± 47	71 ± 25	877 ± 352	Florou <i>et al.</i> (1992)
Taiwan	30	44	431	Yu-Ming <i>et al.</i> (1987)
Canary Islands	44	54	665	Fernandez <i>et al.</i> (1992)
Spain	13-165	7-204	48-1570	Baeza <i>et al.</i> (1992)
Italy	57-71	73-87	580-760	Bellia <i>et al.</i> (1997)
Spain	20.3-711	13.2-84.4	289-703	Martinez-Aguirre <i>et al.</i> (1997)
Russaifa, Jordan	48.3-523.2	8.7-27.1	44-344	Al-Jundi (2002)
Gudalore, India	37.7 ± 10.1	75.3 ± 44.1	195.2 ± 85.1	Selvasekarapandian <i>et al.</i> (2000)
Orissa, India	350 ± 20	2825 ± 50	180 ± 25	Mohanty <i>et al.</i> (2004)
Jaduguda, India	50.67-109.14	28.73-89.78	494.84-1121.36	Present study

TABLE IV
Reported values of gamma activity (activity Bq/kg) of fallout radionuclide ^{137}Cs , from work conducted worldwide.
Valeurs publiées de l'activité gamma (activité en Bq/kg) du radionucléide ^{137}Cs des retombées, dans des recherches effectuées dans le monde entier.

Region	^{137}Cs	Reference
Serbia and Montenegro	48.3 ± 26.19	Dragović <i>et al.</i> (2006)
Antarctica	1.59-15.6	Godoy <i>et al.</i> (1998)
Jordan	7.5-576	Hamarneh <i>et al.</i> (2003)
Buyuk menderes basin, Turkey	2.81-20.75	Aslani <i>et al.</i> (2003)
Israel	1.5-20	Lavi <i>et al.</i> (2006)
Venezuela	3.5-15	La Brecque <i>et al.</i> (1992)
Poland	0.0-101.61	Poreba <i>et al.</i> (2003)

concentrations of the anthropogenic radionuclide ^{137}Cs obtained from the present study are compared to studies carried out worldwide. It is seen that the data from the current study lies in the same range as the worldwide data.

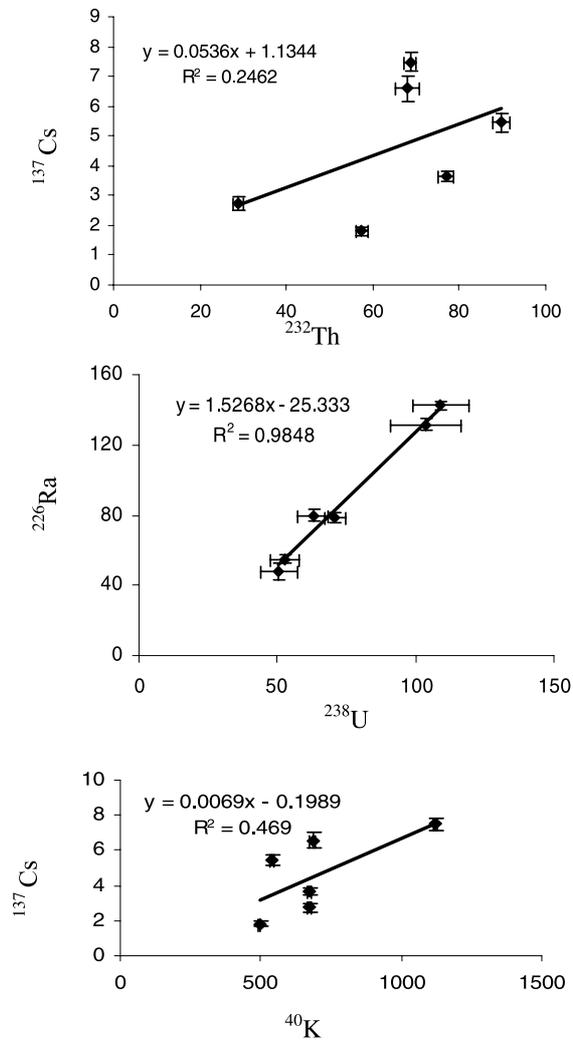


Figure 1 – Correlation of activity concentrations, in Bq/kg, of radionuclides.
Corrélation des concentrations de l'activité (Bq/kg) de radionucléides.

^{238}U , ^{226}Ra , ^{232}Th , ^{137}Cs and ^{40}K activities in the samples were used to investigate whether these radionuclides are correlated. It was found that ^{232}Th has no correlation with ^{226}Ra , ^{40}K and ^{137}Cs . ^{226}Ra and ^{137}Cs show no correlation. ^{238}U shows no correlation with ^{232}Th , ^{137}Cs and ^{40}K . ^{40}K and ^{226}Ra show no

correlation, whereas ^{40}K and ^{137}Cs are positively correlated (correlation coefficient 0.71). A similar trend is evident from Figure 1 which shows the correlation between selected radionuclides. From the figure it is seen that ^{238}U and ^{226}Ra are positively correlated. ^{226}Ra is a daughter product in the natural radioactive decay series of ^{238}U . Hence the correlation of the two radionuclides is expected. ^{137}Cs and ^{40}K though of different origins, the former anthropogenic and the latter naturally occurring, have similar properties, as they are both alkali metals. Hence their behaviour in the environment can be similar, leading to the very weak correlation observed between the two radionuclides. ^{238}U does not show any correlation to other naturally occurring radionuclides, ^{232}Th and ^{40}K , and the fallout radionuclide ^{137}Cs due to their different physico-chemical properties. Similar results have been reported (Al-Jundi, 2002) for NORMS occurring in soil samples collected near an old phosphate mine.

Th/U ratio is indicative of the relative depletion or enrichment of the radioisotopes. Concentrations of ^{238}U and ^{232}Th in the samples ranged from 4.1 to 8.8 ppm and 7.1 to 22.2 ppm, respectively. The concentration ratio (in ppm) of $^{232}\text{Th}/^{238}\text{U}$ is 0.85, 1.61 and 3.3 for the samples 1, 5 and 2 respectively and varied from 3.87 to 4.47 for samples 3, 4 and 6. For the samples 1, 2 and 5 the ratio $^{232}\text{Th}/^{238}\text{U}$ is less than the Clark's value (3.5) indicating uranium enrichment at these locations, whereas at the other locations the ratio is greater than 3.5, denoting no preferential uranium enrichment (El-Dine, 2008).

3.1. Radiation hazard

3.1.1. Radium equivalent activity (Ra_{eq})

An index called the radium equivalent activity (Ra_{eq}) is used to assess the exposure to radiation (Bahari *et al.*, 2007; Beretka and Mathew, 1985; El-Arabi, 2005; Nada, 2003). It is calculated by the following equation:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

where A_{Th} , A_{Ra} and A_K are the activity concentration (Bq/kg) of ^{232}Th , ^{226}Ra and ^{40}K respectively. The radium equivalent concept allows the use of a single index to describe the gamma output from different mixtures of ^{232}Th , ^{226}Ra and ^{40}K in any sample (Bahari *et al.*, 2007).

Equation (1) shows that 1 Bq/kg of ^{226}Ra , 0.7 Bq/kg of ^{232}Th or 13 Bq/kg of ^{40}K will generate the same gamma ray dose rate (Chowdhury *et al.*, 1999). The Ra_{eq} for the soil samples are given in Table V. Values range from 216-263 Bq/kg,

TABLE V
Radiation hazard indices associated with the soil samples.
Indices de risque associés aux divers échantillons de sol.

Sample code	D (nGy/h)	H _{ex} (Bq/kg)	H _{in} (Bq/kg)	ARa _{eq} (Bq/kg)
1	106 ± 8	0.61	0.96	224
2	107 ± 11	0.62	0.84	230
3	113 ± 7	0.67	0.88	248
4	100 ± 7	0.58	0.73	216
5	121 ± 5	0.71	1.0	263
6	111 ± 8	0.63	0.76	233

which are below the internationally accepted value of 370 Bq/kg (Beretka and Mathew, 1985; El-Arabi, 2005). This shows that the radiological hazard for the samples is below the internationally accepted values considering the Ra_{eq} hazard index.

3.1.2. External hazard index (H_{ex})

The external hazard index (H_{ex}) due to the gamma rays emitted from the samples is also used to assess the radiation hazard and is given by the equation (Beretka and Mathew, 1985; El-Dine, 2008; Nada, 2003):

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810). \quad (2)$$

H_{ex} values for the soil samples are all below unity as given in Table V. This indicates that these soils pose no hazard to the workers and peasants in this region.

3.1.3. Internal hazard index (H_{in})

The internal exposure due to ²²²Rn and its daughter radionuclides present in the soils (outdoor conditions) can be indicated by the internal hazard index (H_{in}), which is given by (Nada, 2003):

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_K/4810). \quad (3)$$

From Table V, it is evident that H_{ex} values for the soil samples are below unity. This indicates that these soils pose no hazard to the workers and peasants in this region, considering the H_{in} values.

TABLE VI
Reported values of gamma absorbed dose rates (nGy/h), from work conducted worldwide.
Valeurs publiées des débits de dose absorbée gamma (nGy/h), dans des recherches effectuées dans le monde entier.

Region	Gamma dose rate (nGy/h)	Reference
Rajasthan, India	67 (mean)	Nageswara <i>et al.</i> (1996)
Istanbul, Turkey	65 (mean)	Karahan <i>et al.</i> (2000)
Taiwan	62 (mean)	Yu-Ming <i>et al.</i> (1987)
Canary islands	67 (mean)	Fernandez <i>et al.</i> (1992)
Greece	69 (mean)	Probonas <i>et al.</i> (1993)
Kerala, India	200-4000	Sunta <i>et al.</i> (1981)
Ooty, India	31.6-221	Selvasekarapandian <i>et al.</i> (1999)
Gudalore, India	74.3 ± 27.8	Selvasekarapandian <i>et al.</i> (2000)
Namibia	90-180	Steinhausler <i>et al.</i> (1992)
Russaifa, Jordan	97.5	Al-Jundi (2002)
Red sea coast, Egypt	26.5-50.9	El-Arabi (2005)
Orissa, India	1925 ± 718 (mean)	Mohanty <i>et al.</i> (2004)
World range	28-120	UNSCEAR (1993)
Jaduguda, India	100-121	Present study

3.1.4. Air absorbed gamma dose rate (D)

The total air absorbed gamma dose rate (nGy/h) due to the mean activity concentrations of Ra, Th and K (Bq/kg) in the samples was calculated as (UNSCEAR, 2000)

$$D = 0.463A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (4)$$

D values for the samples range from 100-121 nGy/h. Values from the current study have been compared to work conducted worldwide in Table VI. The values reported in Orissa, India (Mohanty *et al.*, 2004) are for a High Background Radiation Area. ¹³⁷Cs mean activity concentrations have not been included in the estimation of D values since their values are low. Their inclusion in calculation makes little difference to the calculated dose rate.

The contribution of the ²³⁸U series (²²⁶Ra), ²³²Th series and ⁴⁰K to the calculated gamma dose rate are shown in Figure 2. Contribution of the ²³⁸U series varies from 20.1-57.2%, ²³²Th series from 16.4-48% and ⁴⁰K from 17.0-42.3% in the samples. The high degree of variation in contribution of the radionuclides to

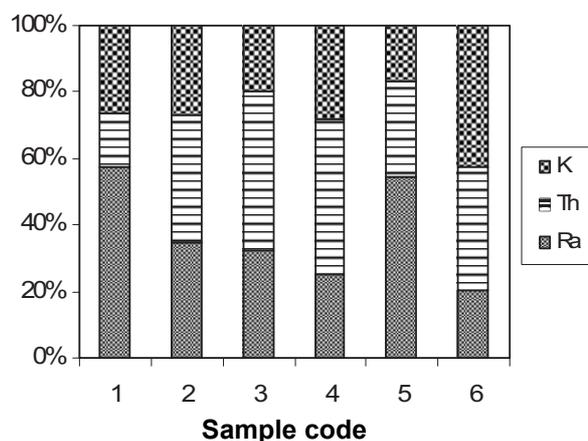


Figure 2 – Contribution of the ^{40}K , ^{232}Th and ^{226}Ra to the calculated dose rate.
Contributions de ^{40}K , ^{232}Th et ^{226}Ra au débit de dose calculé.

dose rate is due to the high variability in radionuclide concentrations in the samples. It is considered that 98% of the external gamma dose rate from ^{238}U series is due to ^{226}Ra and its daughters (Shanbhag *et al.*, 2005). Therefore, any disequilibrium between ^{238}U and ^{226}Ra , will not affect the dose rate estimation due to ^{226}Ra . In the soils around Nilgiri, the estimated contributions to the calculated dose rate have been reported to be (Selvasekarapandian *et al.*, 2001) 66% (^{232}Th series), 20.9% (^{238}U series) and 48.7% (^{40}K). In Indian soils the contribution is as follows, 48.7% (^{40}K), 33.6% (^{232}Th series) and 17.7% (^{238}U series) (Shanbhag *et al.*, 2005). In Indian rock formations the values are 26.7% (^{238}U series), 66.8% (^{232}Th series) and 6.5% (^{40}K) (Sankaran *et al.*, 1986; Nambi *et al.*, 1987). Global values for dose rate estimation from soils were found to be 40% (^{232}Th series), 25% (^{238}U series) and 35% (^{40}K) (UNSCEAR, 2000).

Calculated gamma dose rate, obtained from the activities of ^{226}Ra , ^{232}Th and ^{40}K in the samples was compared to the dose rate in air measured by a portable digital dose rate gamma survey meter (target FieldSPEC). The observed dose rate, varied from 120-200 nGy/h.

Parametric and non-parametric tests have been used in the present study to study the difference between the observed and the calculated gamma dose rates. The Wilcoxon rank sum/Man-Whitney test (non-parametric test) was employed to compare the medians of the calculated dose rates and the observed dose rates. One of the test statistics calculated (1 and 35) was much lower than the critical value (13 for 6 no. of samples). This indicated that at the given level of significance the

OCCURRENCES OF NORMS AND ¹³⁷CS IN SOILS

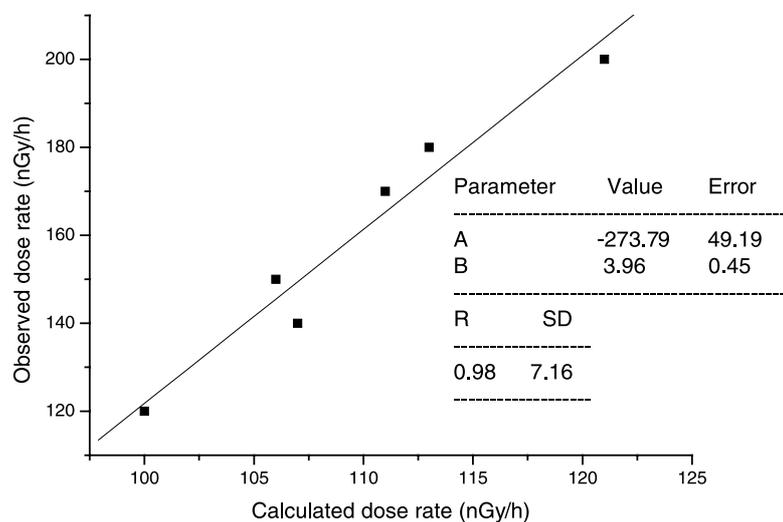


Figure 3 – Linear regression of Pearson correlation coefficient.
Régression linéaire du coefficient de corrélation de Pearson.

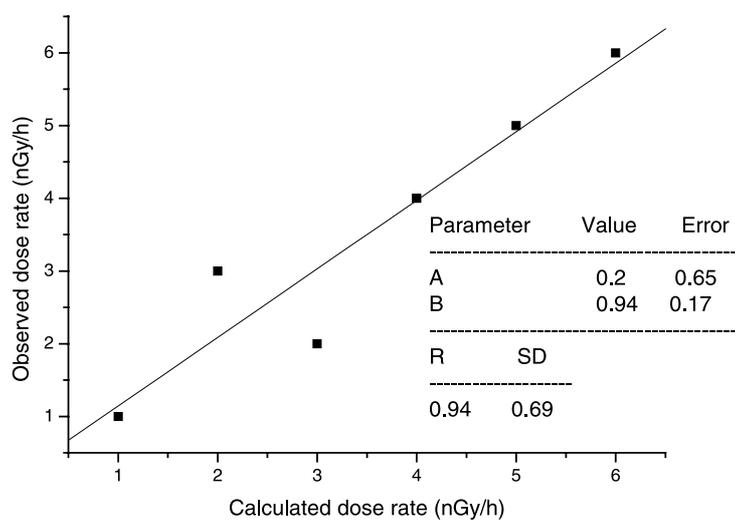


Figure 4 – Linear regression of Spearman's rank (ρ) correlation coefficient.
Régression linéaire du coefficient de corrélation de la classe de Spearman (ρ).

medians of the observed and calculated dose rate are significantly different (Kumar *et al.*, 2007). Pearson correlation coefficient (parametric test) was calculated for the observed and calculated dose rates and its linear regression is shown in Figure 3. The corresponding non-parametric test, Spearman's rank (ρ) correlation coefficient was also calculated and its linear regression is shown in Figure 4. Pearson correlation coefficient gives coefficient of $R = +0.98$ and intercept of -273.79 . Spearman's rank (ρ) correlation coefficient gives $R = +0.94$ and intercept of 0.2 . In both the cases the calculated and observed dose rates show a high correlation. From these tests it is inferred that the observed outdoor gamma dose rate may be influenced by other factors apart from the calculated dose rate, computed using the radionuclide activities, which may be the cause of the difference between the two sets of values. The calculated dose rate is an estimate of the dose rate, at 1m level, due only to the radionuclides present in the soil at that location. It does not take into contribution the cosmic dose rate present at the location. This component may be the reason for the observed difference in the computed and measured dose rates.

4. Conclusion

High Resolution Gamma Spectrometry system was used to study the distribution of NORMS and fallout ^{137}Cs present in surface soils in the Singhbhum region of Eastern India. The activity concentrations varied from 50.67-109.14 Bq/kg (^{238}U), 48.12-142.55 Bq/kg (^{226}Ra), 28.73-89.78 Bq/kg (^{232}Th), 494.84-1121.36 Bq/kg (^{40}K) and 1.8-7.48 Bq/kg (^{137}Cs). The activity concentrations of ^{238}U , ^{232}Th and ^{40}K were found to be slightly higher than global averages. This was due to natural uranium mineralization occurring in this region.

Correlation between radionuclides was studied. ^{137}Cs was seen to have a positive correlation with ^{40}K (+0.71 correlation coefficient). Th/U concentration ratio (Clark value) was also calculated and uranium enrichment was found in some of the soils.

Radiation hazard for the samples was assessed by radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}) and absorbed gamma dose rate (D). The values are below global averages, indicating no radiation hazard to the people in this region.

Calculated and observed dose rates in air were compared with the help of parametric and non-parametric tests like Wilcoxon rank sum/Man-Whitney test (non-parametric test), Pearson correlation coefficient (parametric test) and Spearman's rank (ρ) correlation coefficient. The tests showed difference between the calculated and observed dose rates, indicating that the observed

outdoor gamma dose rate may be influenced by other factors apart from the calculated dose rate.

Acknowledgement. *The authors are thankful to Shri. H.S. Kushwaha, Director HS&E Group for his guidance and to the authorities of UCIL for extending necessary facilities during the course of this work. The authors also acknowledge the encouragement and support received from their colleagues.*

REFERENCES

- Ahmad N. *et al.* (1997) Indoor radon levels and natural radioactivity in Jordanian soil, *Radiat. Prot. Dosim.* **71**, 231-233.
- Al-Jundi J. (2002) Population doses from terrestrial gamma exposure in areas near to old phosphate mine, Russaifa, Jordan, *Radiat. Meas.* **35**, 23-28.
- Aslani A.A.M. *et al.* (2003) Activity concentration of caesium-137 in agricultural soils, *J. Environ. Radioact.* **65**, 131-145.
- Baeza A. *et al.* (1992) Natural radioactivity in soils in the province of Caceres (Spain), *Radiat. Prot. Dosim.* **45** (1/4), 261-263.
- Bahari I. *et al.* (2007) Radioactivity and radiological risk associated with effluent sediment containing technologically enhanced naturally occurring radioactive materials in amang (tin tailings) processing industry, *J. Environ. Radioact.* **95**(2-3), 161-170.
- Bellia S. *et al.* (1997) Natural radioactivity in a volcanic island Ustica, Southern Italy, *Appl. Radiat. Isot.* **48**, 287-293.
- Beretka J., Mathew J. (1985) Natural radioactivity of Australian building materials. Industrial wastes and by-products, *Health Phys.* **48**, 87-95.
- Chowdhury M.I. *et al.* (1999) Distribution of radionuclides in the river sediments and coastal soils of Chittagaong, Bangladesh and evaluation of the radiation hazard, *Appl. Rad. Isot.* **51**, 747-755.
- Dikshitulu G.R. *et al.* (1997) Uranium mineralization at Mouldih, Singhbhum shear zone, Bihar – An ore petrological study, *J. At. Min. Sci.* **5**, 81-86.
- Dragović S. *et al.* (2006) Classification of soil samples according to geographic origin using gamma-ray spectrometry and principal component analysis, *J. Environ. Radioact.* **89**, 150-158.
- El-Arabi A.M. (2005) Natural radioactivity in sand used in thermal therapy at the Red Sea Coast, *J. Environ. Radioact.* **81**, 11-19.
- El-Dine N.W. (2008) Study of natural radioactivity and the state of radioactive disequilibrium in U-series for rock samples, North Eastern Desert, Egypt, *Appl. Radiat. Isot.* **66**, 80-85.
- Faure G. (1986) *Principles of Isotope Geology*, 2nd Ed. Wiley, New York.
- Fernandez J.C. *et al.* (1992) Natural radiation in Tenerife (Canary Islands), *Radiat. Prot. Dosim.* **45**(1/4), 545-548.
- Florou H. *et al.* (1992) Gamma radiation measurements and dose rates in the coastal areas of a volcanic island, Aegean Sea, Greece, *Radiat. Prot. Dosim.* **45**(1/4), 277-279.
- Godoy *et al.* (1998) ^{137}Cs , ^{226}Ra , ^{210}Pb and ^{40}K Concentrations in Antarctic Soil, Sediment and Selected Moss and Lichen Samples, *J. Environ. Radioact.* **41**(1), 33-45.
- Hamarneh I.A. *et al.* (2003) Radioactivity concentrations of ^{40}K , ^{134}Cs , ^{137}Cs , ^{90}Sr , ^{241}Am , ^{238}Pu and $^{239+240}\text{Pu}$ radionuclides in Jordanian soil samples, *J. Environ. Radioact.* **67**, 53-67.
- Higgy R.H. (2002) Radioactivity in sediment and sea-water used in climatotherapy in Safaga, Red Sea, Egypt. In: Sixth Radiation Phys. Conf. Arab Journal of Nuclear Science and Applications, 581-587.
- Karahan G. *et al.* (2000) Assessment of gamma dose rates around Istanbul, *J. Environ. Radioact.* **47**, 213-221.

- Kumar A. *et al.* (2007) A non-parametric statistical analysis in the measurement of outdoor gamma exposure to the residents around Trombay, *Radiat. Prot. Dosim.* **124**(4), 378-384.
- La Brecque *et al.* (1992) The preliminary results of the measurements of environmental levels of ^{40}K and ^{137}Cs in Venezuela, *Nucl. Instr. Meth. A* **312**, 217-222.
- Lavi N. *et al.* (2006) Monitoring and surveillance of radio-caesium in cultivated soils and foodstuff samples in Israel 18 years after the Chernobyl disaster, *Radiat. Meas.* **46**, 78-83.
- Martinez-Aguirre A. *et al.* (1997) Radioactivity impact of phosphate ore processing in a wet marshland in southwestern Spain, *J. Environ. Radioact.* **34**, 45-57.
- Ménager M.T. *et al.* (1993) Migration of uranium from uranium-mineralised fractures into the rock matrix in granite: implications for radionuclide transport around a radioactive waste repository. In: Fourth International Conference of Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere (Migration 1993), Charleston, USA, 12-17 December 1993, *Radiochim. Acta* **66/67**, 47-83.
- Mohanty A.K. *et al.* (2004) Natural radioactivity in the newly discovered high background radiation area on the eastern coast of Orissa, India, *Appl. Radiat. Isot.* **38**(2), 153-165.
- Nada A. (2003) Evaluation of natural radionuclides at Um-Greifat area, eastern desert of Egypt. *Appl. Radiat. Isot.* **58**(2), 275-280.
- Nageswara M.V. *et al.* (1996) Natural radioactivity in soil and radiation levels of Rajasthan, *Radiat. Prot. Dosim.* **63**(3), 631-642.
- Nambi K.S.V. *et al.* (1987) Country-wide Environmental Radiation Monitoring using thermoluminescent Dosimeters, *Radiat. Prot. Dosim.* **18**, 31-38.
- Poręba G. *et al.* (2003) Concentration and vertical distribution of ^{137}Cs in agricultural and undisturbed soils from Chechło and Czarnocin areas, *Geochronometria* **22**, 67-72.
- Probonas M. *et al.* (1993) The exposure of the Greek population to natural gamma radiation of terrestrial origin, *Radiat. Protect. Dosim.* **46**(2), 123-126.
- Sankaran A.V. *et al.* (1986) U, Th and K Distributions Inferred From Regional Geology and the Terrestrial Radiation Profiles in India, BARC Report.
- Selvasekarapandian S. *et al.* (1999) Gamma radiation dose from radionuclides in soil samples of Udagamandalam (OOTY) in India, *Radiat. Prot. Dosim.* **82**(3), 225-228.
- Selvasekarapandian S. *et al.* (2000) Natural radionuclide distribution in soils of Gudalore, India, *Appl. Radiat. Isot.* **52**(2), 299-306.
- Selvasekarapandian S. (2001) Background radiation survey of the Nilgiris Biosphere of Peninsular India, Final report of the DAE/BRNS sponsored project, 1995-1999, Coimbatore, April, 2001.
- Shanbhag A.A. *et al.* (2005) Natural radioactivity content in beach sands of Ratnagiri coast, Maharashtra, *Environm. Geochem.* **8**(1-2), 304-308.
- Steinhausler S. *et al.* (1992) Radiometric survey in Namibia, *Radiat. Prot. Dosim.* **45**(1/4), 553-555.
- Sunta C.M. *et al.* (1981) Analysis of dosimetry data of high natural radioactivity areas of SW coast of India. In: Vohra, K.G. (Ed.), *Proc. Natural Radiation Environment*, New Delhi, 35-42.
- Tzortzis M. *et al.* (2004) Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus, *J. Environ. Radioact.* **77**, 325-338.
- UNSCEAR (1993) Sources, effects and risks of ionizing radiation. Report to the General Assembly, with Scientific Annexes. UN, New York.
- UNSCEAR (2000) Sources and Effects of Ionising Radiation, United Nations. Report to General Assembly with Scientific Annexes. United Nations, New York.
- Yu-Ming L. *et al.* (1987) Measurement of terrestrial gamma radiation in Taiwan, Republic of China, *Health Phys.* **52**, 805-811.