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ACKNOWLEDGEMENTS. I thank the Department of Science and Technology, New Delhi for providing financial support through a project (No. DST/ESS/16/301/2006). I also thank Rajesh Srivastava, Banaras Hindu University, Varanasi for giving me an opportunity to report the discovery of these dykes at the 6th International Dyke Conference at Varanasi, and the anonymous reviewers for their suggestions and critical comments.

Received 1 March 2014; revised accepted 22 April 2015

Distribution of naturally occurring radionuclides uranium and ^{226}Ra in groundwater adjoining uranium complex of Turamdih, Jharkhand, India

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Estimation of radionuclide content is essential for assessment of individual exposure in areas where groundwater is the principal source of drinking water. Elevated levels can be expected in areas known for radioactive mineral deposits and anthropogenic activities like mining and ore processing industry. The aim of this study is to determine the uranium and ^{226}Ra in groundwater sources adjoining and away from uranium mining and ore processing industry at Turamdih, Jharkhand. The concentration of uranium in well/tubewell samples analysed nearby and away from the tailings ponds ranged from 0.1 to 8.4 μg l^{-1} and ^{226}Ra varied from 4 to 269 mBq l^{-1}. The wide variation of activity concentration is due to regions of uranium deposits with elevated level of radium in the earth's crust and geological faults, when compared to lower concentration profile of radium in earth crust. The ingestion of uranium and ^{226}Ra in the adult population residing around Turamdih mining complex through drinking water sources ranged from 0.81 μSv year^{-1} to 3.8 μSv year^{-1} respectively. This is much lower than 100 μSv year^{-1}, that is recommended by WHO for ingestion from intake of a single radionuclide. The groundwater monitoring carried out over four years around Turamdih mining complex indicates that there has been no observable impact on groundwater sources due to mining and ore processing activities in this region.

Keywords: Groundwater, ingestion dose, ^{226}Ra, uranium.

PRESENCE of naturally occurring radionuclides in groundwater is a significant source of background radiation exposure[1,2]. In areas, where groundwater is the principal source of drinking water, estimation of radionuclide content is essential for assessment of an individual's internal exposure. Wide variation in radionuclide level in groundwater is observed depending on the geological features of the concerned area and other environmental variables. Low concentration of the radionuclide is invariably present in most of the environmental compartments including drinking water[3]. Elevated levels can be expected in areas known for radioactive mineral deposits. Apart from these natural deposits, anthropogenic activities such as mining and processing of minerals may also contribute significantly in enhancement of the radionuclide levels in groundwater[4–7]. Earlier studies[8,9] in areas of uranium mining and ore processing suggest that the concerned radioniclides from environmental protection point of view are uranium (U) and ^{226}Ra. Moreover, these radionuclides are also significant source of natural radiation exposure. Both U and ^{226}Ra have large radiological half lives (4.5×10^9 year and 1620 year respectively) and metabolic interactions with living beings. Apart from the radiological concern, U is also chemically toxic[2]. ^{226}Ra, owing to its similarity in chemical properties with calcium and emission of high-energy alpha particles followed by generation of radioactive decay products, its interactions with the metabolic system of living beings is anticipated[10]. Unlike other mining and processing industry,

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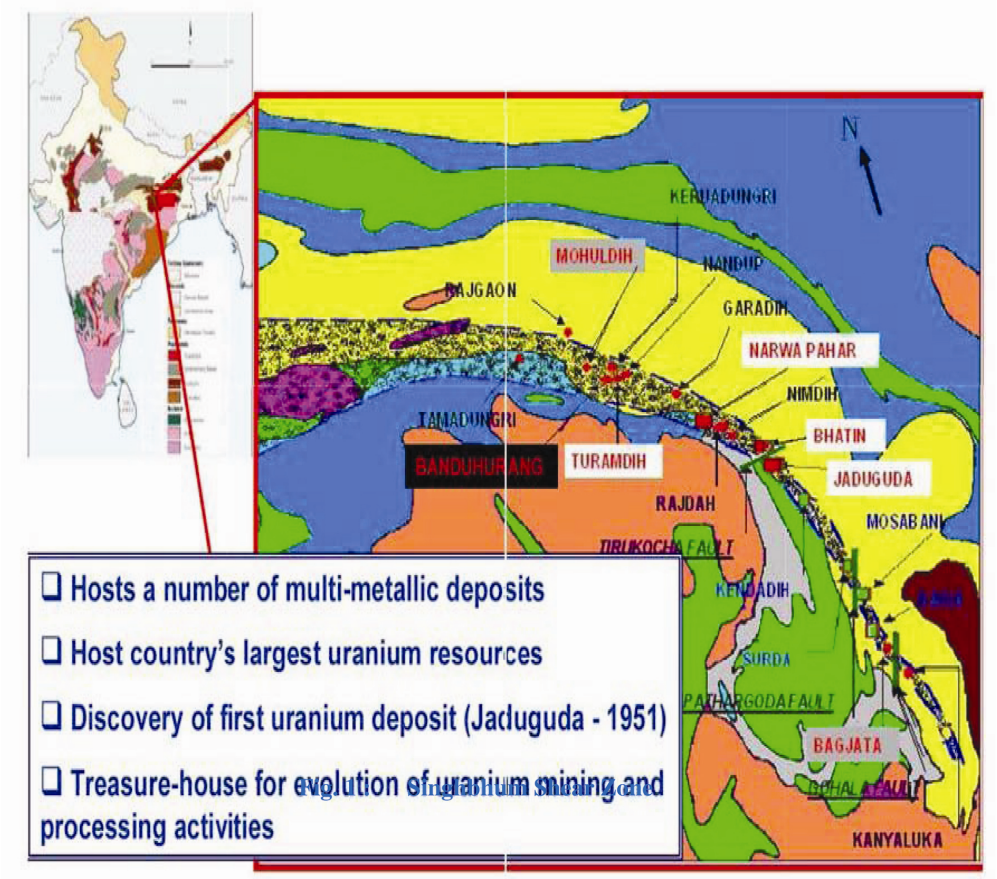

Figure 1. Singhbhum shear zone.

uranium mining and processing industry produces a large quantity of low specific waste in the form of slurry called tailings. Tailings is subsequently impounded, preferably in a natural valley with a hill barrier and earthen embankment. Migration of contaminants from the tailings impoundment facility is a probable source of groundwater contamination[11]. Singhbhum region of Jharkhand is known for its widespread mineral deposits. Economically viable grades of uranium and copper have been reported in this area[12,13]. Mining and processing of uranium ore in the Sighbhum shear zone of Jharkhand is carried out for the last five decades. New mining sites have been opened up within a distance of about 25 km from the existing Jaduguda facilities. The ore deposits are at Turamdih, Mohuldih and Banduhurang mines, out of which the first two mines are underground and the third one is the country's first open cast uranium mine. A new ore processing mill with a capacity to process 3000 tonne per day has been commissioned during the year 2007 at Turamdih near these ore deposits to process the ore from all the three mines. The mined ore is subjected to sulphuric acid leaching using pyrolusite oxidant, followed by ion exchange separation and product recovery in the form of magnesium di urinate (MDU). Large quantity of solid and liquid waste is generated during the processing of uranium ore. The solid waste is generated from the filtration unit and the liquid waste is barren liquor from the ion exchange column. These two wastes are mixed and neutralized using lime at an elevated pH (>9). The neutralized waste (tailings) is separated into coarse (35%) and fines (65%) fractions, the coarse fraction is sent to the mines for back filling and fines are discharged in an engineered impoundment system called the tailings pond[9]. At the tailings pond, fine solids are settled and liquid is decanted out for removal of the dissolved radionuclides (U, ^{226}Ra) prior to its discharge into the environment based on the existing national regulatory compliances[14]. The activity of ^{210}Po in the decanted effluent is appreciably low and is attributed to the fixation of Po on $Fe(OH)_3$ and MnO_2, produced during the neutralization of waste. Hydroxide of ^{230}Th is highly insoluble and its presence in the tailings effluent is unlikely. The present study is confined to the distribution of U and ^{226}Ra radionuclides around and away from the impoundment facility. The study incorporates detection of the two radionuclides in groundwater sources in areas with widespread uranium mineralization and uranium industry. Based on the statistical database, ingestion dose due to U and ^{226}Ra to the adult population (adult) residing around and away from Turamdih mining complex is evaluated.

A map of the study area (Singhbhum Shear Zone) is presented in Figure 1. The study was carried out in Singhbhum shear zone of Jharkhand at Turamdih, Mahuldih and Banduhurang areas. Samples were collected from the areas adjoining the mining, ore processing and tailings impoundment facilities. Tailings disposal system at Turamdih is designed based on the latest engineering methods and is located in a natural stable environment, which includes a variety of critical control features such as embankment, linings, covers and water control structures like spill way, drainage system, etc. Turamdih tailings pond is located between two hills running almost parallel towards the south of the process plant. The area is a sloping valley. Downstream of the tailings pond (western side), a decant water pond of 30,000 m^3 has been constructed for storage and pumping the decanted water to an effluent treatment plant (ETP). The decanted effluent is further treated at a centralized effluent treatment plant at Turamdih to ensure long-term environmental protection[15].

A comprehensive environmental monitoring was carried out around Turamdih complex to ascertain the migration of radionuclides into the adjoining groundwater sources, from the tailings pile. Ten bore wells were constructed on the earthen bund across the tailings pond to assess up-gradient and down-gradient groundwater quality. Monitoring of the adjoining groundwater source as well as sources far away from the uranium industry was carried out on a routine basis.

Groundwater samples were collected from individual bore holes constructed near the tailings pond in preconditioned carboys each month. Groundwater samples collected around Turamdih complex were divided into three categories based on their distance, i.e. <1.6 km, 1.6 km to 5 km and >5 km from the tailing pond. Samples were collected from the tube wells and wells used by the local people from around the uranium mining complex. Samples from Turamdih were collected for comparison. Collection of samples was based on the proximity of the source from the sites and its consumption by the local inhabitants. Immediately after collection, the samples were filtered using a 0.45 μm membrane filter paper at the laboratory. Filtered samples were preserved in dilute nitric acid to avoid wall deposition losses. Sampling methodology used by earlier workers was followed[16–18]. All reagents used were of electronic grade. With each set of sample, blank samples containing only the reagents were also tested.

Natural uranium was estimated using combined methods of solvent extraction followed by flourimetry. A known volume of sample was evaporated to dryness using concentrated sulphuric acid for the removal of interferences. The evaporated samples were refluxed in 0.25 N H_2SO_4 and uranium was separated by solvent extraction technique using alamine in a benzene solvent[19]. Along with the samples, NIST standards were also processed in an identical manner. Chemically separated uranium was transferred onto a platinum disc, fused with fusion mixture $NaF–Na_2CO_3$ (15 : 85) and then subjected to UV radiation in a fluorimeter to measure the fluorescence intensity. The uranium content of the original sample was obtained from fluorimetric reading of standard, collected and blank samples by further applying the sampling parameters.

^{226}Ra was estimated by allowing the build up of its daughter ^{222}Rn for a known period (>2 weeks). Concentrated water samples were loaded into a standard air tight radon bubbler. The in-built radon was collected in a scintillation cell by vacuum transfer and counted after equilibrium (between radon and its progeny) was achieved[10]. Initially ^{222}Rn that was already present in the solution was removed by using a vacuum pump. After ensuring the sample was radon free, the solution was retained for two weeks or more depending on the expected level of ^{226}Ra in the sample. Alpha decay of ^{226}Ra leads to the formation and accumulation of its progeny ^{222}Rn, during the retention period. The in-built radon was collected in a previously evacuated scintillation cell. The scintillation cell was left for a minimum of 200 minutes to ensure equilibrium of ^{222}Rn and its short lived progeny. The scintillation cell was counted for alpha activity after this delay period. Based on the integrated alpha counts and other decay and sampling parameters, ^{226}Ra was estimated at 95% confidence level using the formula

$$^{226}\mathrm{Ra(Bq)} = \frac{1.883 \times 10^{-3} \times C \times 0.037}{E \times (1 - e^{-\lambda\theta}) \times (1 - e^{-\lambda T}) \times e^{-\lambda t}},$$

where C is the net counts obtained after subtraction of the background, E the efficiency of the cell (85%), t the counting delay in minutes, T the counting duration in minutes, θ the build up period in minutes, λ is the decay constant of ^{222} Rn (1.258×10^{-4} min^{-1}).

Minimum detectable concentration of uranium using the above method works out to be 0.1 μg l^{-1}. The minimum detectable ^{226}Ra activity in the sample solution loaded in radon bubbler is affected by factors like build up period, efficiency and background count rate of the Lucas cell, counting duration, etc. The observed background of Lucas cell as 0.5 cpm and efficiency of the counter at 85%, the minimum detectable activity works out to be 6.8 mBq.

The dissolution of radionuclides into groundwater depends on factors like pH, geological formation of the area, redox potential and chemical speciation of the concerned species. Each radioactive decay product has unique chemical characteristics common to the element but differing slightly by isotope. The occurrence of a parent radionuclide in solution does not necessarily indicate the presence of its decay product. Though ^{238}U (parent of ^{226}Ra) tends to be least mobile in oxygen-poor groundwater and

tends to be strongly absorbed onto humic substances, ^{226}Ra is most mobile in oxygen-poor and chloride-rich groundwater with a high concentration of total dissolved solids[20,21]. Solubility of alpha-particle-emitting radionuclides is also enhanced by alpha recoil process during the decay. The energy associated with recoil is 10^4–10^6 times larger than the typical chemical bond energies[22] and can cause atoms on the surface of a grain to recoil directly into water in pore spaces.

Uranium is soluble in hexavalent state and the most probable complex formation is likely the mechanism of its dissolution. Uranium has two geologically significant oxidation states +6 and +4. In its oxidized state, uranium is quite mobile. The uranyl ion UO_2^{++}, forms bi and tricarbonate complexes that move easily through oxidized environments[23,24].

The natural processes, which lead to the release of radium from rock to underground water are sometimes denoted as 'primary migration' in contrast to 'secondary migration' of the released radium in groundwater, mine drainage water and surface water. Studies carried out by Posokov[25] underline the fact that the transition of radium from rock to groundwater is determined by the type of rock, composition of groundwater, characteristics of the water movement, temperature and time interval of contact. Tanner's[26] observations indicate an increased concentration of radium in groundwater enriched in chloride, pointing to the role of chloride ion in the dissolution of radium from the rock. Radium chemically reacts similarly to other divalent alkaline-earth cations such as calcium, strontium and barium. Radium can enter natural water by a number of routes including leaching of uranium ore by ground and precipitation of water, seepage from tailings pond and leaching of radium from waste solids. The major concern in tailings management system is seepage of contaminated water into surface water and groundwater. Water that is retained in tailings after processing can cause excess pore water pressure, which will expel contaminated water, especially by consolidation[27]. Waste management facilities that are left uncovered may be a source of continuous contaminant seepage. In absence of properly addressed control measures, underground migration of radionuclides from the tailings pond may appear in the adjoining groundwater sources. The slurry from uranium mill tailings contains radionuclide of the entire ^{238}U series such as U (nat.), ^{226}Ra, ^{230}Th, ^{210}Po. The likelihood of migration of ^{230}Th and ^{210}Po is extremely low, the former due to the insoluble nature of hydroxide and the latter due to the adsorption on $Fe(OH)_3$ and MnO_2 coating as discussed above. Due to the presence of clayey soil and hard rock at the bottom of tailings containment system, the hydraulic conductivity is low[9,28].

The analytical results of 178 samples for U and ^{226}Ra and the median concentration are presented in Figures 2 and 3. The concentration of U in the bore wells around the tailings pond ranges from 0.11 μg l^{-1} to 2.44 μg l^{-1} with a geometric mean (GM) of 0.98 μg l^{-1} and a geometric standard deviation (GSD) of 1.2, while activity concentration of ^{226}Ra varies from 3 mBq l^{-1} to 22 mBq l^{-1} with its geometric mean at 8.3 and the geometric standard deviation at 1.2. The maximum concentration of U at 2.44 μg l^{-1} was found in bore well no. 4 and is far below the EPA primary standard of 30 μg l^{-1} for uranium in drinking water. For ^{226}Ra, the maximum value was found to be 22 mBq l^{-1} in bore well no. 9, which is only 12% of the maximum contaminant level (MCL) for combined ^{226}Ra and ^{228}Ra in drinking water, which is 185 mBq l^{-1} as specified by the USEPA[29]. The typical concentration of U in tailings effluent varies from 50 to 100 μg l^{-1}, whereas for ^{226}Ra it varies from 1000 to 3000 mBq l^{-1}.

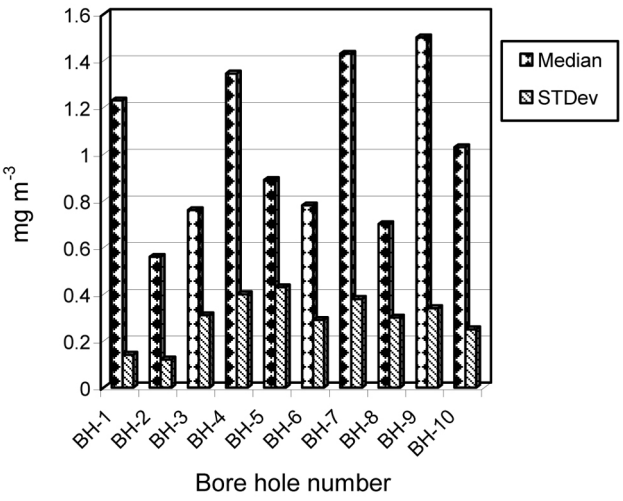

Figure 2. Median concentration of U in bore holes around tailings pond.

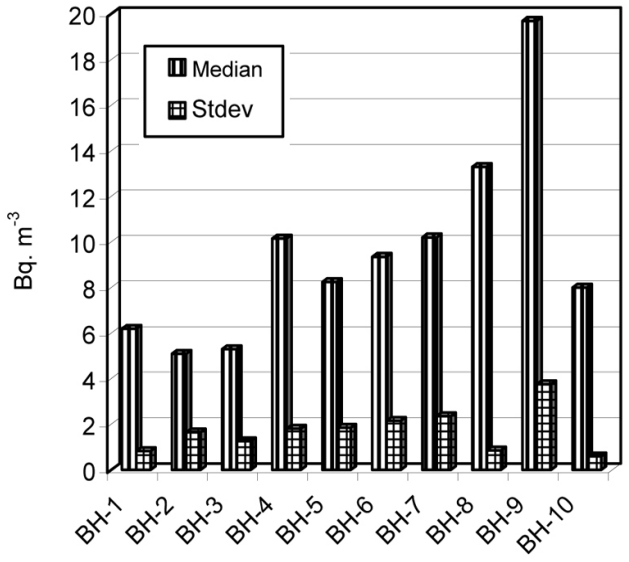

Figure 3. Median concentration of ^{226}Ra in bore holes around tailings pond.

The levels of both these radionuclides are significantly lower than those observed in the tailings effluent which may be attributed to local geological features of the area. Migration of contaminants has not been observed in the area so far. These levels are comparable with the groundwater sources situated in the vicinity of the tailings pond and adjacent areas within the same geological formation.

Results of 153 samples of groundwater (well and tube well) collected regularly over a period of four years after commissioning of the facility was analysed. The frequency distribution, mean, standard deviation, skewness and kurtosis were evaluated for each data set in each sector. No data set from all the sectors follow a normal distribution, either for uranium or for radium. These data sets were further analysed for the test of log normality and results of the statistical analysis are discussed. Based on the Jarque-Bera test of normality, uranium's natural distribution at <1.6 km follows a log normal distribution, with $h = 0$ and $p = 0.05$. The GM and GSD can be taken as unbiased estimators. Concentration of uranium in the villages near to the tailings pond (<1.6 km) varied from 0.1 μg l^{-1} to 5.1 μg l^{-1} with a GM of 1.3 μg l^{-1} and a GSD of 1.4. In villages of the second sector (1.6–5 km from tailings pond) uranium concentration varied from 0.1 to 2.6 μg l^{-1} with a GM of 0.8 μg l^{-1} and GSD of 1.3. The data set obtained in this sector follows a log normal distribution, based on Lilliefors test with $h = 0$ and $p = 0.06$. The median value (0.8 μg l^{-1}) of the data and GM are identical, which also confirms the log normal trend. Concentration of uranium at distances far away from tailings pond (>5 km) ranges from 0.2 to 8.4 μg l^{-1}. The values observed in this sector neither follow normal nor log normal distribution, based on JBKS (Jarque-Bera and Komogorov-Smirnonov) test and Lilliefors test. The frequency distribution of U is presented in Figure 4. A data set does not have a definite distribution, median and percentile values have been evaluated. The 95% confidence interval and the median is 0.2 μg l^{-1} and 8 μg l^{-1} respectively. The 5th and 95th percentile are 0.2 μg l^{-1} and 6.7 μg l^{-1} respectively. The maximum likelihood estimate (MLE) value is 1.3 μg l^{-1} and 1.8 μg l^{-1}. At a MLE of 1.3 μg l^{-1} confidence intervals varied from 0.72 μg l^{-1} to 1.97 μg l^{-1}, while at MLE of 1.8 μg l^{-1} confidence interval ranged between 1.4 μg l^{-1} and 2.35 μg l^{-1}. Data from all the three sectors for a two-way analysis of variance reveal that there is no variation ($p = 0.45$) in the concentration between the years, while there is slight variation within the distance ($p = 0.03$). The maximum concentration of uranium (8.4 μg l^{-1}) is observed at a distance greater than five km away from the tailings pond. Drinking water standards of WHO are based on health consideration and are designed to protect human health. The maximum value observed in the present study is much below than drinking water guidelines of WHO that are either based on chemical toxicity (15 μg l^{-1}) or radiological toxicity (400 μg l^{-1}).

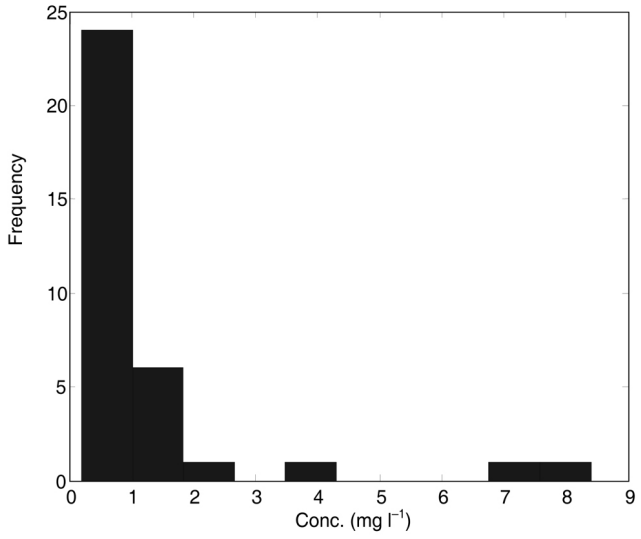

Figure 4. Frequency distribution of U (nat.) at a distance of >5 km.

^{226}Ra concentration in the sector near to tailings pond (<1.6 km) varies from 5 to 269 mBq l^{-1} with a GM of 20.3 mBq l^{-1} and a GSD at 1.5. The median and the GM of the data are 19.5 mBq l^{-1} and 20.3 mBq l^{-1} respectively. Though slightly skewed the lognormal approximation is valid for this data set. The concentration of ^{226}Ra in the second sector (1.6–5 km) varied from 5 to 99 mBq l^{-1} with a GM of 19.8 mBq l^{-1} and the GSD at 1.4. In this sector, some of the data were censored (MDL) and only 4 data points (out of 66) are outliers with respect to median (16.5 mBq l^{-1}). At distances far away from the tailings pond (>5 km), concentration of ^{226}Ra varied from 4 to 163 mBq l^{-1} with a GM of 18.1 and the GSD at 1.5. Large deviation from log normality was found in this sector. The MLE concentration was evaluated to be 33 mBq l^{-1} and 44.5 mBq l^{-1}. At MLE of 33 mBq l^{-1}, the 95% confidence interval (CI) was between 17.2 mBq l^{-1} and 48.7 mBq l^{-1} and at a MLE of 44.5 mBq l^{-1}, the 95% confidence interval ranged from 36.4 to 59.5 mBq l^{-1}. Two-way analysis of variance of total data in all the sectors over the years shows that there is neither a variation between the distance nor the year (between the distance $p = 0.90$, within the year $p = 0.88$). The activity concentration of ^{226}Ra in the groundwater is well below the guidelines value of 1000 mBq l^{-1} (ref. 30). Out of 154 samples analysed, only 6 samples have the activity concentration more than 100 mBq l^{-1}. The reason for wide variation of activity concentration is due to regions of uranium deposits with elevated level of radium in the earth's crust and geological faults with lower concentration profile of radium in the earth's crust[11].

Well and tube well water are the principal sources of drinking water in this region. Ingestion dose can be assessed through drinking pathway for adult population using the yearly intake of drinking water and dose

conversion factor for both the radionuclides. The health effects of uranium in drinking water are chronic (the delayed result of continuous consumption over a large period of time). Mass concentration of natural uranium observed in groundwater can be expressed in activity concentration using a conversion factor of 25 Bq mg^{-1}. Taking the GM of uranium concentration of all data sets, the activity concentration deduced to be 24.8 Bq m^{-3}.

Ingestion dose can be computed using the formula

$$D\ (\mu Sv\ year^{-1}) = \text{Water intake}\ (m^3\ year^{-1}) \times \text{GM activity conc.} \times \text{dose conversion factor}\ (Sv\ Bq^{-1}).$$

The intake of uranium and radium by people is calculated assuming 2 l day^{-1} of intake[31]. Applying the dose conversion factor of 0.045 μSv Bq^{-1} (refs 32, 33) the ingestion dose due to U for the adult population around Turamdih complex is 0.81 μSv $year^{-1}$. This value is appreciably lower than the recommended guidelines of 100 μSv $year^{-1}$ (ref. 28) for ingestion from intake of a single radionuclide.

Radium concentrations in bone when ingested and exposure to ^{224}Ra, ^{226}Ra and ^{228}Ra are associated with bone carcinoma at quite elevated level. Ingestion dose to adult population due to ^{226}Ra was calculated based on the geometric mean value of the observed data. Ingestion dose is estimated using BSS[32] dose conversion factor from intake of ^{226}Ra (Sv Bq^{-1}) in the above formula. The annual estimated ingestion dose due to ^{226}Ra from intake of water in the region was 3.8 μSv $year^{-1}$, which is about 94% lower than the WHO recommended values.

The groundwater monitoring carried out for over four years around Turamdih mining complex indicates that there has been no observable impact on groundwater sources due to the mining and ore processing activities in this region. The wide variation of U and ^{226}Ra concentration in groundwater is attributed to geological formation and local mineralization of uranium in this area. The ingestion dose due to U and ^{226}Ra to adult population residing around Turamdih mining complex from drinking water sources is 0.81 μSv $year^{-1}$ and 3.8 μSv $year^{-1}$ respectively. Both these values are far lower than the WHO recommended guidelines of 100 μSv $year^{-1}$ (ref. 30) for ingestion from intake of a single radionuclide. The study reveals that various control measures taken at every stage of uranium mining ore processing and tailings management are effective in restricting the migration of contaminants from tailings pile to the adjoining groundwater sources.

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ACKNOWLEDGEMENTS. We thank Dr Pradeep Kumar, Associate Director, Health Safety and Environment Group, BARC, Mumbai for his keen interest and guidance during this study. We also thank the authorities of Uranium Corporation of India Limited for providing necessary facilities. The help and support received by the colleagues of Health Physics Unit, Turamdih, Jaduguda and Narwapahar is also acknowledged.

Received 26 February 2015; accepted 31 March 2015