Assessment of chemo-radiological risk of naturally occurred uranium in groundwater from the Beed district, India

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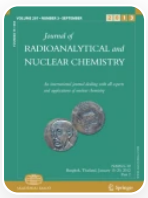

Journal of Radioanalytical and Nuclear Chemistry

Aims and scope

Submit manuscript

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Abstract

The main objective of study is assessment of chemo-radiological risk using measured concentrations of uranium in drinking water. For estimation of U concentration, LED Fluorimeter was used. For data accuracy, statistical tools were applied, geographical distribution GIS based software were used. To assess relation between uranium to other parameters, correlation test was performed. On the basis of presence of U in the water, hazard quotient, effective dose and cumulative dose for lifetime were estimated, it ranges

from 0.001 to 1.09, 0.01–18.61 μSv year^{-1} and 1.22–1303 μSv for lifetime correspondingly. The estimated ingested dose is well below than the suggested limit of 0.1 mSv.

Introduction

In drinking water contain radioactive substances that might present a risk to human health [1]. In recent centuries, there has been increasing concern over uranium (U) in drinking water due to its chemo toxic as well as radiotoxic property. The important objects that affect in an individual body are tubular cells of kidney, lungs [2] and bone [3] effects with various form of cancer [4] due to intake of uranium contaminated drinking water. U is categorized as (group A) i.e. carcinogenic element recommended by USEPA in 1991. Also, it is suggested that, the absolute non-existence of uranium in drinking water as the safe limit for carcinogenic risk. In recent the United States Environmental Protection Agency (USEPA) and World Health Organisation (WHO) has planned the guideline for maximum contaminant Level (MCL) for uranium is 30 μg L^{-1} [1, 5]. The chemical toxicity is a chief challenging health effect from uranium, rather than radiological risks [1, 6]. In groundwater, U mainly form complexes with phosphate and carbonates, but at lower pH, it has an affinity towards fluorides and chloride and their

complexes [7] The uranium is the basic key component in production of nuclear power. It is an atomic number 92, metal having in silvery-white in the actinide series in the periodic table. This naturally occurs in rock, soil and water. It is having three isotopes i.e. U^{238} with half-life 4.5×10^9 years, the Second one is ^{235}U with half-life-700 Million years and the third one is ^{234}U with half Life-244,000 years correspondingly. Uranium is undergone series of deterioration and the final stable product is Lead (^{206}Pb) [8]. The World Health Organization had previously recommended a reference level 15 µg L^{-1} for U, but at present, its permissible limit is 30 µg L^{-1} in drinking water [9], and as per guidelines of Indian Atomic Energy Regulatory Board, it is 60 µg L^{-1} [10] U in natural groundwater system depends upon several aspects such as lithology, geomorphology, and other geological aspects of the region. Moreover, the spatial deviation of U generally depends on geochemical factors (rock-water interaction) and its residence time in groundwater [11] Even though its toxic property, the uranium is highly toxic and it is not normally measured as an indicator of drinking water quality. The main purpose of the present study is to measure the level of uranium in drinking water samples as well as assess the water quality parameters and it's become very significant to estimate the radiological as well as chemo toxicological risk, due to the absorption of uranium from the point of view of health risk.

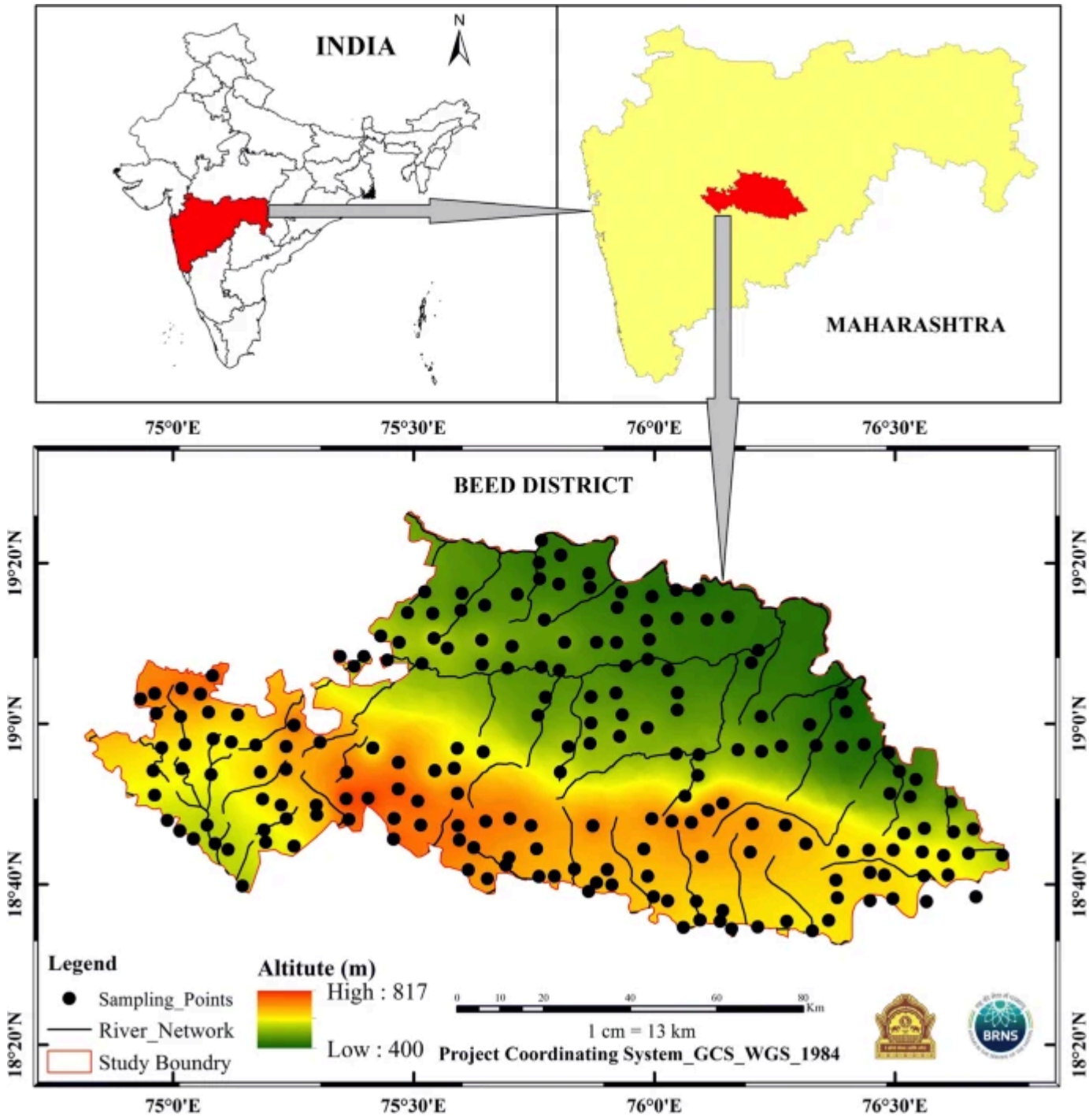
Experimental

Hydrogeology of study area

Beed district is positioned in the central portion of Maharashtra in Aurangabad division and also a part of Marathwada region. The study area as shown in (Fig. 1) is lies between 18° 27′ and 19° 27′ North Latitudes and 74° 49′ and 76° 44′ East Longitudes. Study area is situated towards North side from Aurangabad and Jalna Districts; by Parbhani and Latur to the East; by Osmanabad and Ahmadnagar Districts to the South; and by Ahmadnagar district to the West site. Balaghat Hill ranges route through the district, these hill ranges are about 600–670 m above main sea level. North part of the study area falls under the Godavari Valley, whereas the Southern part falls under the Manjra river valley. The population of Beed district was 25.86 lakh (Census 2011). It covers a geographical area of 10,963 km^2. The deposition of alluvial in the river courses of Godavari and Sindphana in Sub-recent age are observed along. The mean annual rainfall for the district is 750.1 mm. About 80% of the annual rainfall is received in the south-west monsoon period. September is the wettest month. The variation in the rainfall from year to year is fairly large. All the streams of the district drain into one of the three principal rivers viz., the

Godavari, the Manjra and the Sina. The Godavari River is the main river which runs through the Northern part of the district as well as Manjra, Sindphana, Bindusara and Wan are other rivers in the district. There are several small rivers in the district, which go dry during summer season [12].

Fig. 1



Showing the study area

Sampling

The 200 number of water samples were collected across the Beed district with making the 6 × 6 km grid map. The random sampling method was chosen for the collection of samples based on population density shown Fig No. 1 Before the collections of water samples, the air-tight lab grade polypropylene bottles having capacity 1 L were washed with 10% hydrochloric acid (HCl) to remove impurities that are immersed to the interior surface of the bottle. Fresh water samples were taken by running bored well for 2–3 min prior collection to ensure that the fresh water is sampled from the aquifer. For the minimization for contamination in the collection of water sample, filtration, storage, and handling, standard protocol has been followed [13]. For the accurateness of the results, the Physico-chemical parameters i.e. Temperature, pH, EC, TDS, Dissolved Oxygen, ORP and Salinity were analysed on the sampling spot with the help of Portable Multiparameter Meter kit (Orion Star A326). For uranium analysis, the samples are labelled, representing the particulars of time, place and date of sampling. The concentration level of uranium was analysed in the departmental laboratory in University.

Determination of physiochemical parameters

The in situ parameters i.e. [pH, oxidation–reduction potential (ORP), Electrical conductivity, Dissolved Oxygen, Total Dissolved Solids (TDS), salinity, and temperature] were analysed on site with the help of Thermo Scientific Orion Star A326 Portable Multiparameter Meter kit. The certain anions in water, phosphate, nitrate, and sulphate were determined following the Stannous Chloride Method, Screening Method, and Turbidimetry Method respectively using a UV spectrophotometer (Bio Era Single Beam UV–visible spectrophotometer). The chloride ions were determined using Mohr's method. For total hardness, calcium and magnesium were estimated using a standard EDTA titration method. All procedures followed for analysis were as per standard methods for the examination of water and wastewater [14].

Uranium estimation

The naturally presented uranium concentration in the collected water samples were analysed with the help of LED Fluorimeter (Quantalas LF-2a). The LED Fluorimeter is working on the principal of fluorescence of uranyl ion [14] The Fluorescence value varies for different complexes of uranium. Therefore, all the complexes were converted into a single form having same fluorescence yield, by addition of (5%) sodium pyrophosphate ($Na_4P_2O_7{\cdot}10H_2O$) solution as a Fluorescence enhancing reagent (Fluren) were used. The

doubled distilled water was used to prepare the Fluren reagent. To adjust the 7.00 pH, 10% phosphoric acid is added drop wise. The LED fluorimeter was standardized with the help of standard samples containing 1, 3, 5, 7, and 10 µg L^{-1} of uranium alongside observed values of fluorescence.

Methodology of risk assessment

In the present study, the risk was assessed in two types separately is related to the occurrence of uranium in drinking water. The principal one is a radiological risk which is due to the ionizing radiations emitted by radioactive element uranium and another one is chemical risk. The main radiation exposure, occurs when uranium complexes are ingested or inhaled by the individual's body. Uranium is a carcinogenic heavy metal and it is injurious to human health. The most important chemical effect related to exposure of uranium is kidney toxicity. So, it is become very significant to estimate the risks associated with uranium.

Radiological risk (ECR) is evaluated using the following equation

$$\begin{aligned} {\text{Radiological risk (ECR)}} &= {\text{conc}} . {\text{ of U in groundwater}}{\mkern 1mu} ({\text{Bq L}}^{ - 1}){*} \\ &\quad\times {\text{risk factor (per}}{\mkern 1mu} {\text{Bq L}}^{ - 1}){**} \end{aligned}$$

(1)

$$\begin{aligned} {*{\text{Conc}}} . {\text{ of U }}({\text{Bq L}}^{{ -^{1} }}) &= {\text{analysed value of U}}\,({\text{mg L}}^{{ -^{1} }})\\ & \quad\times {\text{conversion factor }}(0.025{\mkern 1mu} {\text{Bq L}}^{{ -^{1} }}) \\ {** {\text{Risk factor}}} &= {\text{risk coefficient }}({\text{Bq L}}^{{ -^{1} }}) \\ &\quad\times {\text{water ingestion rate }}({\text{L day}}^{{ -^{1} }}) \\ & \quad \times \,{\text{total exposure duration day}} \\ \end{aligned}$$

(2)

Average daily dose (LADD) and this can be assessed by using the following equation

$$\begin{aligned} &{\text{Lifetime Average Daily Dose}}\, [({\text{LADD}}),\upmu{\text{g kg}}^{{ - {\text{day}}}}] \\ &= \frac{{[{\text{Cd}}}

$$\times {\text{IR}} \times {\text{EF}} \times {\text{LE}}]}}{{[{\text{BW}} \times {\text{AT}}]}} \end{aligned}$$

(3)

$${\text{HQ}} = \frac{{({\text{LADD}})}}{{({\text{Rfd}})}}$$

(4)

Results and discussion

Analysis of results

The concentration of uranium from 200 different locations groundwater samples collected within Beed districts is presented statistically in Table 1. It varies from as 0.03 to 32.85 µg L^{-1} with an average value is observed 2.58 µg L^{-1} and a median 1.40 µg L^{-1}. Since median is significantly lower than mean value which is consistent with high positive skewness, representing that there were some high values. The groundwater in the study area is observed as chemically of Ca−HCO_3 type with the approximately high amount of TDS (3rd quantile 800.7 mg L^{-1}), which is above the desirable limit (500 mg L^{-1}) at slightly towards alkaline pH (7.50) (Table 1) [15]. An experiential value of EC (> 3000 µS cm^{-1}) was also found in some of the samples in this region but most of them were found above the permissible limits [16]. Observed DO (4.3–9.8 mg L^{-1}) and ORP (18.1–216.9 mV) values ranged between oxic to the suboxic condition. The detected concentrations of nitrate are ranged in between 10 and 300 mg L^{-1} is clearly indicate pollution by anthropogenic activity like the use of fertilizers. The level of U concentration in groundwater samples varied from 0.03 to 32.85 µg L^{-1} with an average of 2.58 µg L^{-1} and most of the samples have U level less than permissible values recommended by the internationally accredited agencies such as AERB, USEPA shown in Table 2, except one is cross the limit which is suggested by WHO i.e. 30 µg L^{-1} at the RAMGAON (19.12N, 75.94) and concentration is 32.85 µg L^{-1}.

Table 1 Statistics of dissolved natural uranium content and other physicochemical parameters

Table 2 Permissible limit of uranium conc. in drinking water of different duthority

Statistical relationship

A Pearson's correlation matrix was built for assessing the dependency of variables (i.e., concentration of several water quality parameters) using statistical software SPSS version 23. The correlation matrix of uranium with other parameters is represented in Table 3. The significantly correlation was observed between U and further parameters [22]. A strong positive correlation coefficient (0.36) with p value < 0.01 was observed among U and electrical conductivity (Table 3). This finding suggests that the existence of U was moderately related with the background water composition, sedimentary rocks beneath the sub surface layer, and mineral-augmented groundwater. Subsequently, this statistical conclusion with uranium may imply the weathering and suspension process of minerals in the vadoze zone or might be surplus consumption of groundwater rather than anthropogenic sources. The pre-eminent U constituents (or precipitate) relate to these primary and/or secondary minerals, which may be unconstrained to the groundwater by roll-front U deposit actions. The reacting state (positive ORP value and DO) can also help to release and transport of U in aquifers [16].

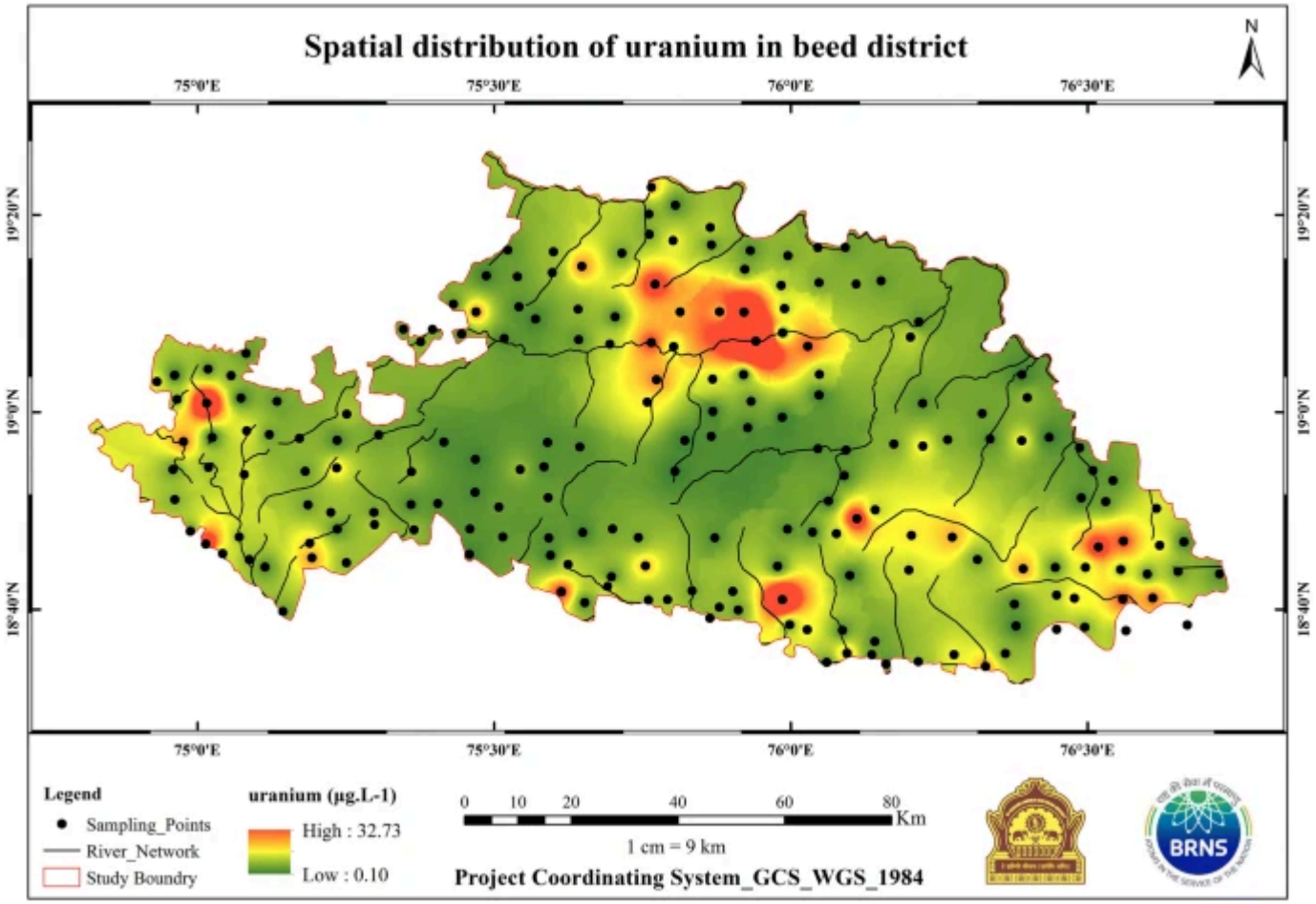
Table 3 Correlation coefficient matrix of uranium with other physicochemical parameters

Spatial distribution of U and its associated parameters

For predicting the unkown concentration of uranium form the konwn sample points of the study area, the spatial analyst modelling tool (ArcGIS 10.3) was used. The method is used for creating maps is interpolation (IWD) fixed-length method. Inverse distance weighted (IDW) interpolation method is determines the values of points close to sampled points were more probable to be related than those farther apart. These types of methods are helpful for risk assessment based on spatial variability and finding the sample spots. The inverse distance weighting interpolation for forecasting Uranium as well as it's correlation with water quality parameters for conclusion the outline of their distribution

after generating raster surface were applied. The resulting maps were shown an elevated level of U was found only in one spot, Fig. 2. In the case of chloride, nitrate, sulphate, phosphate, TDS, EC, salinity, Total Alkalinity and Total Hardness towards increased from southern part towards northern part of study area. The spatial distribution of uranium is showing in the Fig. 2 and remaining parameters distribution are attached in supplementary documentory.

Fig. 2



Spatial distribution of uranium in study area

Radiological risk evaluation

The risk coefficient in Eq. (2) for mortality and morbidity was taken as 1.19×10^{-9} Bq^{-1} and 1.84×10^{-9} Bq^{-1} respectively. The rate for water ingestion was taken as 1.38 L^{-1} and total contact period was taken 25,550 days. Calculated risk of mortality and morbidity is calculated and it is 4.19×10^{-5} and 6.48×10^{-5} respectively. The calculated cancer risk for

mortality and morbidity was found in the range from 3.24×10^{-8} to 3.44×10^{-5} with the mean value of 2.7×10^{-6} and in the range from 5.22×10^{-8} to 1.56×10^{-1} with the mean value 7.8×10^{-3} respectively.

Assessment chemical risk

The chemical toxicity risk associated to any component is evaluated in terms of lifetime. Where in Eq. (3), Cd is denoted uranium concentration in groundwater in ($\mu g\ L^{-1}$), Ingestion Rate (IR) in (L^{-day}) is which is taken to be 1.38 L^{-day}. The EF is the Exposure Frequency (Days–Year) which is taken 365 days per year. LE is the life expectancy was taken 70 years. BW is the Body Weight (kg), which is taken as 70 kg. AT is an average time (Days), which is taken as 25,550 days. In Eq. (4) HQ is denoting Hazard Quotient (HQ) and Rfd showing the Reference dose (μg/kg/Day), which is taken as 0.857 μg/kg/Day [17]. The calculated chemical toxicity Risk i.e. Lifetime Average Daily Dose was found in varying in the range from 8.86×10^{-4} to 0.93 (μg/kg/Day) with mean Value of 0.073 (μg/kg/Day). The calculated Hazard Quotient (HQ) was found in between 0.001033 and 1.096, with an average value 0.0899 (Table 4).

Table 4 Statistical parameters of obtained data

Assessment of Annual Effective Dose

The "Annual Effective Dose" is a biophysical dose; it determines how unsafe an individual's exposure to radiations. The unit of 'effective dose' is the Sievert. It was estimated using the conversion factors given by

$${\text{DE}} = {\text{Ac}} \times F \times I({\text{annual}})$$

(5)

where in Eq. (5) is the annual effective dose ($\mu Sv\ year^{-1}$), Ac is the average concentration, *F* is the effective per unit intake ($\mu Sv\ year^{-1}\ Bq\ L^{-1}$), which is taken 4.5×10^{-8} and *I* is the annual ingestion, which was taken 503.7 L (1.38 × 365). The estimated Annual Effective Dose was found in the range from 0.0175 to 18.6154 $\mu Sv\ year^{-1}$ with an Average

1.4614 μSv year^{-1}. The cumulative Dose was calculated for lifetime, and it is found varying in the range from 1.23 to 1303 μSv with the mean value 102.30 μSv. The uranium concentration and estimated radiological and chemical risks related with each water samples are shown in Table 2.

Conclusion

From present estimation, the spatial distribution of uranium in groundwater and assessed possible health risks due to ingestion of water is carried out in Beed district. Large spatial variations (0.03–32.82 μg L^{-1}) of U were observed in drinking water. From the spatial analysis, it was observed that the concentrations of water quality parameters were potentially increasers from southern part to northern part due to the surplus use of fertilizers and may be due to the river basin of Godavari. In the case of uranium, a uniform variation of concentration was also observed throughout the regions with mostly in the range of 0.04–4.15 μg L^{-1}. In the study area, amount of uranium presented in ground water is significantly correlated with TDS, EC, salinity and total hardness with the p value 0.01. Therefore, the U concentration in groundwater should be continuously studied at these positions including the neighbouring areas to investigate the source; that would be the scope of this research. The ingestion dose evaluation, carcinogenic risk (ECR), and non-carcinogenic (chemo-toxicity risk) exposed that the mean radiological as well as chemo-toxicological risk is insignificant and well below the prescribed permissible limits recommended by Atomic Energy Regulatory Board and World Health Organization. The committed effective dose as whole body, due to U intake through drinking water, was found to vary from 0.01 to 18.61 μSv year^{-1}, with an average value 1.47 μSv year^{-1}, which is within the limit i.e. 0.1 mSv recommended by WHO. The mean HQ value (0.086) was observed, which is expressively lower than 1. The calculated lifetime cumulative dose is observed in the range of 1.3–1303 μSv lifetime^{-1} with mean value 102 μSv lifetime^{-1}. On these criteria some samples which are cross the limits recommended by nationally and internationally research institute working which is working on this area theses samples source need to be reconsidering for potable use. If possible, water consumption from sample point should be avoided for drinking purpose.

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Ethics declarations

Conflict of interest

The author’s declare that they have no conflict of interest.

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Supplementary material 1 (DOCX 2124 kb)

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