Spatial and temporal variation of uranium in a shallow weathered rock aquifer in southern India

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Uranium occurs naturally in groundwater and surface water. The objective of this study is to understand the causes for the occurrence of uranium and its spatio-temporal variation in groundwater in a part of Nalgonda district, Andhra Pradesh, south India. Uranium deposits occur in the southeastern part of this area. Groundwater samples were collected from 44 wells every two months from March 2008 to January 2009. The samples were analyzed for pH, ORP and uranium concentration. The uranium concentration in groundwater varies from 0.2 ppb to a maximum of 68 ppb with a mean of 18.5 ppb. About 21.6% of the samples were above the drinking water limit of 30 ppb set by USEPA. The uranium concentration varied with fluctuation in groundwater level, pH and ORP. Uranium concentration in groundwater changes depending on lithology, degree of weathering and rainfall recharge.

1. Introduction

Uranium is a naturally occurring radionuclide in surface and groundwater. Natural uranium is a mixture of three isotopes – ^{238}U, ^{235}U and ^{234}U. Its average abundance in the earth's crust is about 2 ppb. It occurs naturally in rocks and minerals such as granite, lignite, phosphate deposits and in uranium minerals such as uraninite, carnotite and pitchblende. Uranium also occurs in low concentration in all natural waters. Intake of uranium can cause chemical as well as radiological toxicity which usually affects the kidneys (through ingestion from water or food) and the lungs (through inhalation). Zamora *et al* (1998) had reported cases of kidney disorders due to ingestion of drinking water containing uranium as high as 780 ppb. Garshasbi *et al* (2005) reported the uranium concentration in seawater of Caspian Sea and Persian Gulf to be around 6.12 ± 0.18 ppb and 3.53 ± 0.1 ppb. The association of uranium with colloidal and suspended particulate matter in Arabian Sea was studied by Singhal *et al* (2004). The concentration of uranium in various rivers of India such as Yamuna (0.09–3.61 ppb) and Chambal (0.2–1.74 ppb) (Rengarajan *et al* 2006), Bhagirathi (2.11–3.96 ppb) and Alakanda (1.86 ppb) (Sarin *et al* 1992) have been studied earlier. Springs and streams in Himachal Pradesh, India had 0.07 to 4.65 ppb of uranium (Singh *et al* 1999). Similarly uranium concentration in groundwater has been reported in several parts of India – Kumaun and Siwalik (1.08 ± 0.02 to 35.83 ± 0.09 ppb) (Ramola *et al* 1988), Punjab (11.71 ± 0.15 to 113.70 ± 0.46 ppb) (Singh *et al* 1995), Kolar district (0.3 to 1442.9 ppb) (Babu *et al* 2008) and Upper Siwaliks (1.08 ± 0.03 to 19.68 ± 0.12 ppb) (Singh *et al* 2009). The present study was carried out in Nalgonda district, Andhra Pradesh, southern India where occurrence of unconformity related uranium deposits have been reported (Sinha *et al* 1995). The presence of indoor radon/thoron concentration in this area was reported by Reddy *et al* (2003). Singh *et al* (2002) studied the uranium concentration (0.5

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to 410 ppb) in groundwater in Palnadu sub-basin which is located adjacent to the present study area. The causes for the presence of high fluoride concentration in groundwater of parts of Nalgonda district was reported by Brindha *et al* (2011). However, occurrence of uranium in groundwater in this area as well as its spatial and temporal variation has not been studied. Keeping an account on the importance of public health and considering the fact that there is sizable uranium mineralization in this area, this study was aimed to understand the present status of uranium, the causes, spatial and temporal variation in groundwater in Peddagattu region of Andhra Pradesh, southern India.

2. Materials and methods

2.1 *Description of the study area*

The study area forms a part of Nalgonda district, Andhra Pradesh, which is located at a distance of 85 km ESE of Hyderabad (figure 1). The southeastern side of the study area is surrounded by the Nagarjuna Sagar reservoir and the southern side of the area is bounded by Pedda Vagu river. The northern boundary is bounded by Gudipalli Vagu river. This area experiences arid to semi-arid climate. This area goes through hot climate during the summer (March–May) with a temperature ranging from 30° to 46.5°C and in winter (November–January) it varies between 16° and 29°C. The average annual rainfall in this area is about 1000 mm occurring mostly during southwest monsoon (June–September). The topography derived from SRTM (Shuttle Radar Topography Mission) data is shown in figure 2. In general, the ground surface slopes towards southeastern direction. There are several small hillocks in this area with height ranging from 100 to 200 m. The Lambapur Peddagattu area where the uranium minerals occur are flat topped hills with an elevation of about 300 m msl. The major rivers Pedda Vagu and Gudipalli Vagu which forms the two boundaries of the study area are seasonal rivers that flows during the southwest monsoon from July to September. The rainfall has led to dentritic to subdentritic drainage pattern in this area (figure 1). Numerous tanks and few small reservoirs are present in the depressed parts of the undulating topography of the study area. There are also wide lined canal networks catering for irrigation purposes. The forest cover is thin to moderate.

Most of the study area comprises of agricultural land. Paddy is the principle crop grown in this area while other crops include sweet lime, castor, cotton, grams and groundnut. Drip irrigation is practiced in this area especially for sweet lime.

2.2 *Geology*

Granitic rock forms the basement of this region, which is traversed by numerous dolerite dykes and quartz veins (figure 3). Most part of the investigated area has exposures of granitic rocks belonging to late Archaen. Granites are generally medium-to-coarse grained. The Srisailam Formation, the youngest member of the Cuddapah Supergroup uncomformably overlies the basement granite with a distinct unconformity. The Srisailam Formation is exposed in the southeastern part of the

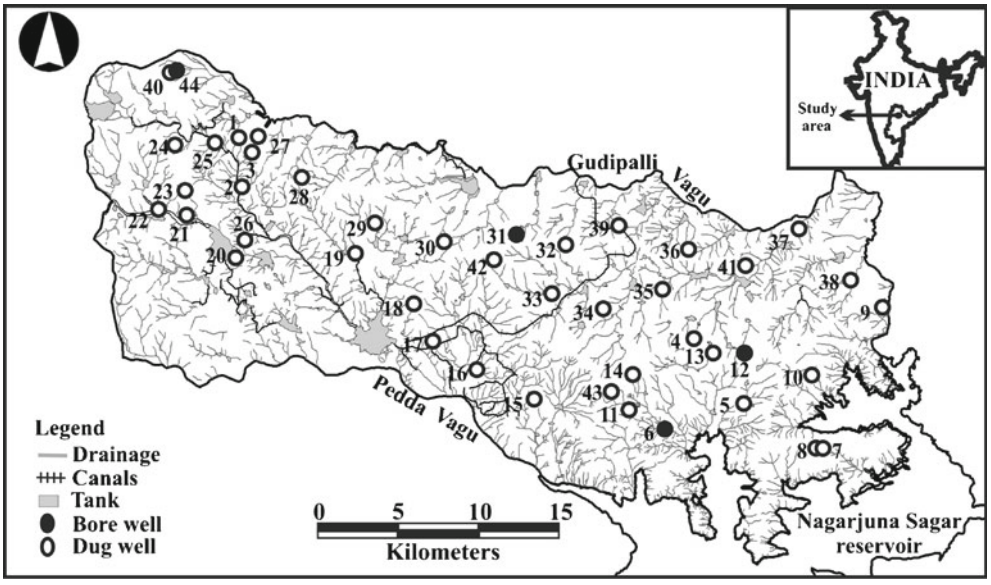

Figure 1. Location of the study area with drainage and monitoring wells.

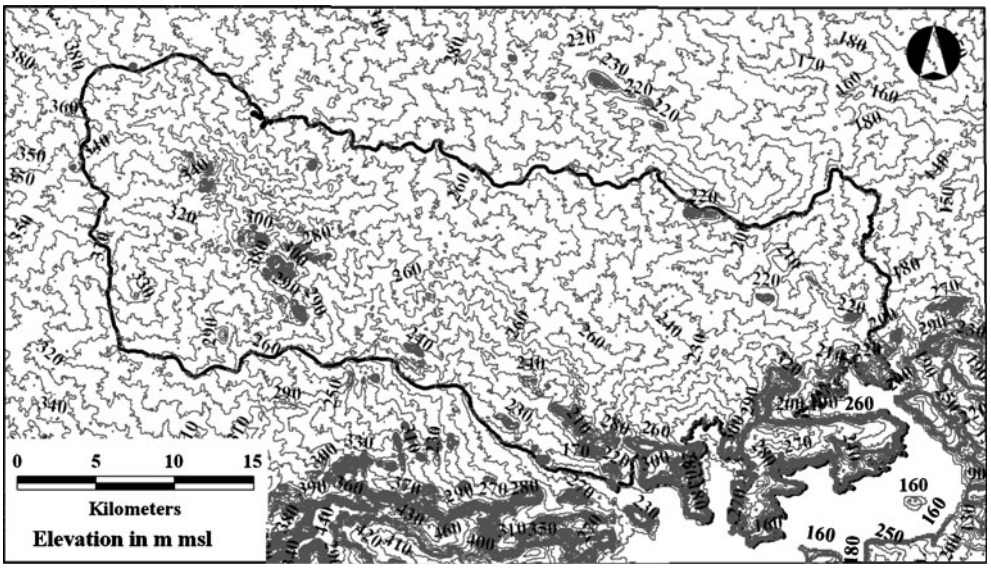

Figure 2. Topography.

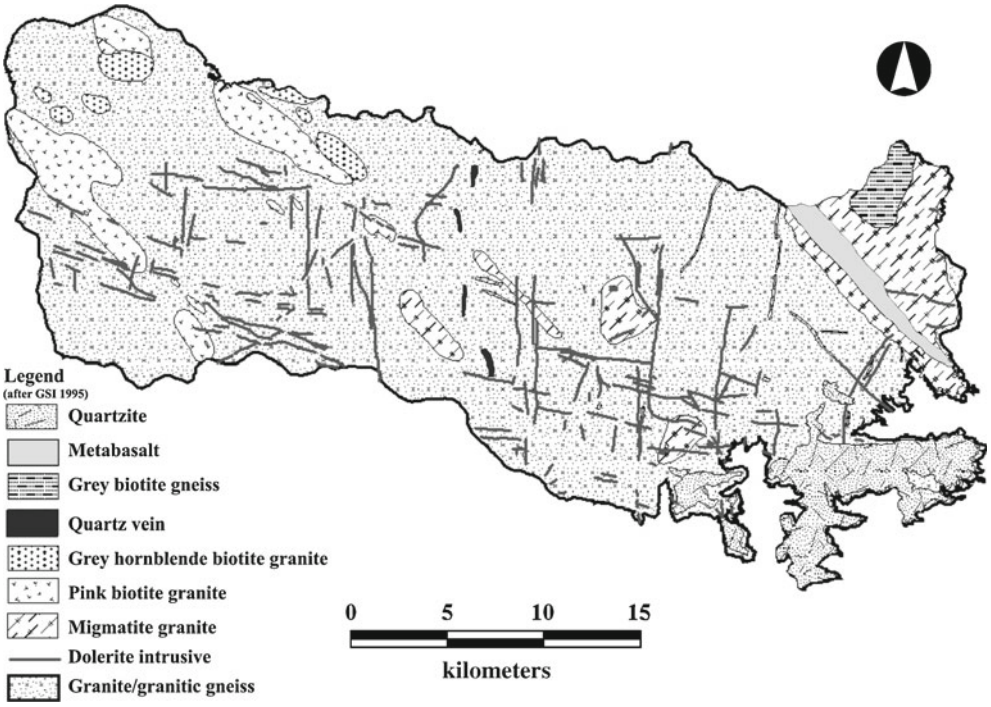

Figure 3. Geology.

study area. The sediments of Srisailam Formation are mainly arenaceous and include pebbly-gritty quartzite shale with dolomitic limestone, intercalated sequence of shale-quartzite and massive quartzite. The litho units of this formation are dipping at an angle ranging from 3° to 5° towards SE. The generalized stratigraphic sequence of this area is given in table 1 (after GSI 1995). The uranium deposit occurs adjacent to the unconformity between basement granite and the overlying Proterozoic Srisailam quartzite in the northwestern margin of the Cuddapah basin. Uraninite, pitchblende, kasolite and uranophane are the main uranium minerals present in the deposits of Lambapur and Peddagattu (Sinha *et al* 1995). The primary uranium mineralization is of epigenetic hydrothermal in nature (Singh *et al* 2002). Lambapur mineralized zone occurs at shallow depth of about 10–15 m whereas it occurs below 50 m or more in Peddagattu. The granitic rocks of this region contain uranium in the range of 10.2–116 ppm with an average of 35 ppm (Shrivastava *et al* 1992).

2.3 *Hydrogeology*

Hydrogeology of the study area was arrived based on field work, monitoring of groundwater levels, pumping tests and infiltration tests. The study area has four distinct layers which are the soil zone, moderately weathered, highly weathered and massive rock. The thickness of soil zone ranges from 0.6 to 12 m. The soil zone is comparatively thicker in the southern and northeastern boundary of the study area due to the influence of rivers. The thickness of the moderately weathered granite ranges from 11 to 77 m and highly weathered granite layer ranges from 12 to 33 m. Most of the wells penetrate up to the fractured layer. The lithology of a few wells are shown in figure 4. The groundwater table occur generally from 0 to 12 m below ground level. The annual groundwater level fluctuation is around 8 m. The principal source of groundwater recharge is rainfall. The groundwater level rises by 1 m after the monsoon rains in the months of July–September. That is, the groundwater levels before monsoon range from 3 to 7 m bgl, and after monsoon, it varies from 2 to 3 m bgl. Groundwater generally flows towards the southeastern direction. Though there are many igneous intrusions in this area, due to the high intensity of weathering and as groundwater occur at shallow depths they do not act as barriers to groundwater flow. The dug wells of this area range from 1.5 to 20 m and dug-cum-bore wells up to 70 m. The yield of the irrigation wells ranges between 100 and 150 m^3/day whereas in few places it is up to 200 m^3/day (CGWB 2007). Most of the wells in this area are used for irrigation purposes. The hydraulic conductivity of the study area generally ranges from 0.5 and 18 m/day. Groundwater plays a predominant role in the net irrigated area by constituting 57.20%, whereas surface water irrigation accounts for 38.63% in Nalgonda district (CGWB 2007).

2.4 *Sampling and instrumentation*

An intensive field survey was carried out and nearly 240 wells were considered for sampling. The EC (electrical conductivity) (μS/cm) of groundwater

Table 1. *Stratigraphic sequence of the study area (after GSI 1995).*

Cuddapah Supergroup	Massive quartzite
Srisailam Formation	Upper shale
	Quartzite with shale intercalation
	Lower shale with limestone intercalation
	Pebbly and gritty quartzite/arenite
	Uranium mineralized region
---------- Unconformity ----------	
Late Archean/Lower Proterozoic	Granite/granitic gneiss with intrusion of dolorite dykes and quartz veins

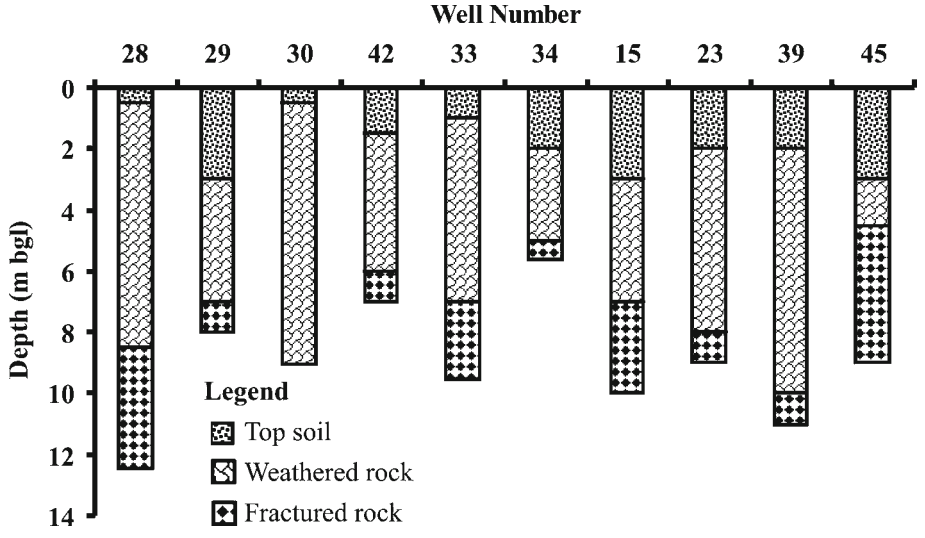

Figure 4. Lithologs of few wells.

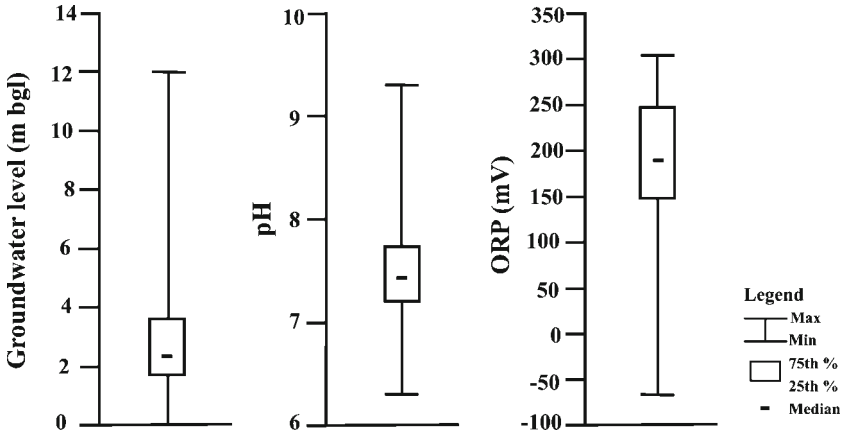

Figure 5. Range and median of parameters measured.

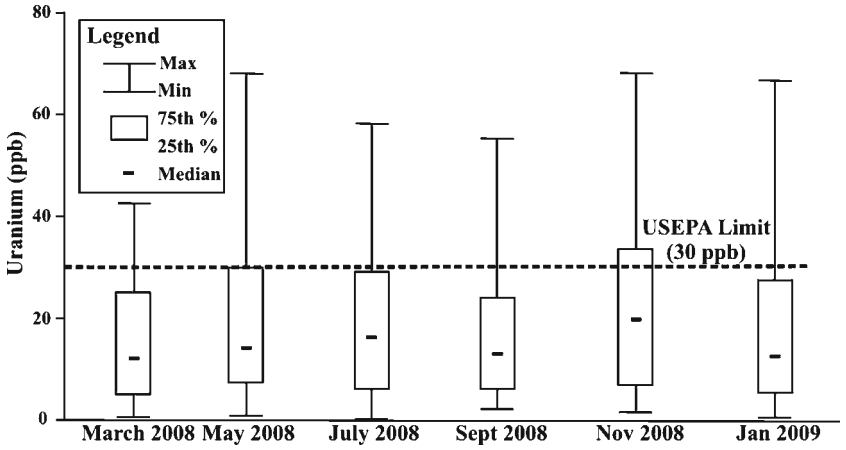

Figure 6. Temporal variation in concentration of uranium in groundwater.

was measured in these wells and a representative well in about every 10 km^2 was chosen for regular collection of groundwater samples. In this way, 44 wells (figure 1) were chosen for the collection of representative groundwater samples. Groundwater samples were collected from these wells once every two months from March 2008 to January 2009. Groundwater level (m), pH and ORP (oxidation reduction potential) (mV) were measured *in situ*. Groundwater level was recorded by using a water level indicator (Solinst 101) while pH and ORP were measured using Eutech digital portable meters. The pH meter was calibrated beforehand using 4.01, 7 and 10.01 buffer solution. Quinhydrone 86 and 255 standard solutions were used to calibrate the ORP meter. All the calibrated solutions and instruments were procured from Eutech Instruments. Groundwater samples were collected in clean polyethylene bottles of 500 ml capacity. The sampling bottles were soaked in 1:1 diluted nitric acid solution for 24 hours, washed with distilled water, and were washed again prior to each sampling with the filtrates of the sample. In the case of bore wells, the water samples were collected after pumping the water for sufficient time so as to collect the formation water. Five types of fertilisers commonly used in this area were also collected. Groundwater samples and fertilisers were analysed for their uranium concentration using laser fluorimeter at Health Physics Unit, Nuclear Fuel Complex, Hyderabad, India. Laser fluorimeter works basically on the fluorescence of a uranyl complex formed during the addition of inorganic complexing regent (sodium pyrophosphate) to the sample during the analysis. Nitrogen laser with a wavelength of 337 nm was used as an excitation source for the determination of uranium concentration in this method (Robbins 1978; Kumar *et al* 2008). Analysis with a sensitivity of about 0.05 ng/g (Shawky *et al* 1994) of uranium can be made

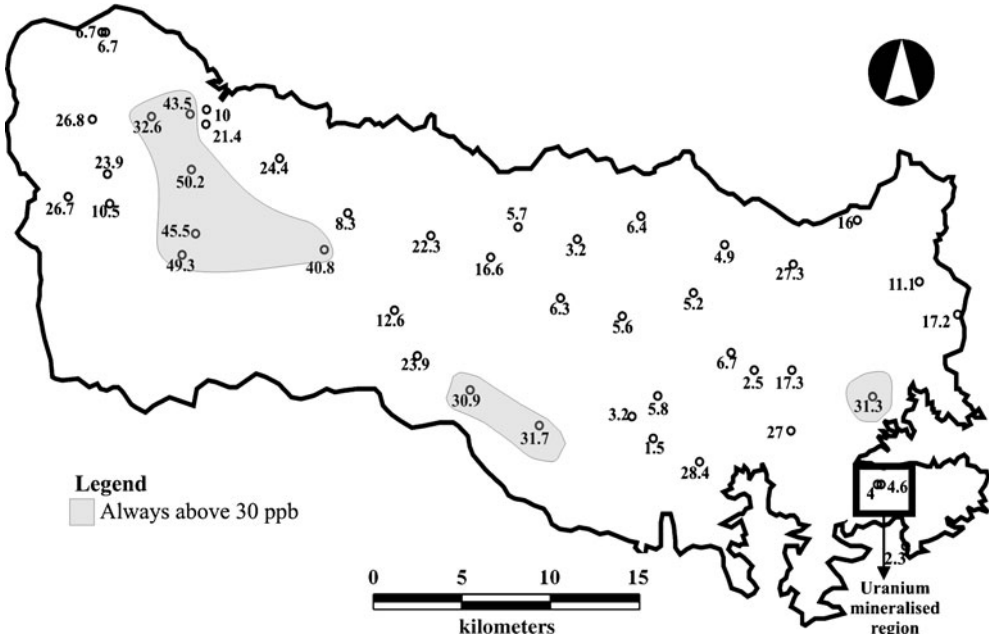

Figure 7. Average uranium concentration (ppb) in groundwater.

without preconcentration or treatment of the sample even in the presence of many potentially interfering species by laser fluorimeter (Sintrex UA-3). Present study used the laser fluorimeter fabricated by Laser Applications and Electronics Division, R.R. Centre for Advanced Technology (CAT), Department of Atomic Energy, Indore, India (Rani and Singh 2006; Kumar *et al* 2008) which has a detection limit of 0.1 ppb. All the analytical reagents used were procured from Merck. Blanks and standards were run simultaneously during the measurement for ensuring accuracy of the result. For every 10 samples, three samples were run in triplicates by varying the concentration of the standard and a calibration curve was obtained to cross check the accuracy of the instrument and to avoid handling errors.

3. Results and discussion

The range of groundwater level and the various physical parameters recorded during the sample collection are shown in figure 5. The statistical summary of the uranium concentration obtained from the analysis of groundwater samples during each sampling is shown in figure 6.

A total of 236 groundwater samples were collected and analysed during this study. The uranium concentration during the study period ranged from 0.2 to 68 ppb. The average uranium concentration recorded in sampling wells and in the reservoir from March 2008 to January 2009 is shown in figure 7. The average concentration of uranium in the groundwater samples was 18.5 ppb. The Indian Standards Specifications for drinking water (BIS 1993) does not specify any maximum permissible limit for uranium. Hence, the USEPA (2003) health standard of 30 ppb of uranium in drinking water

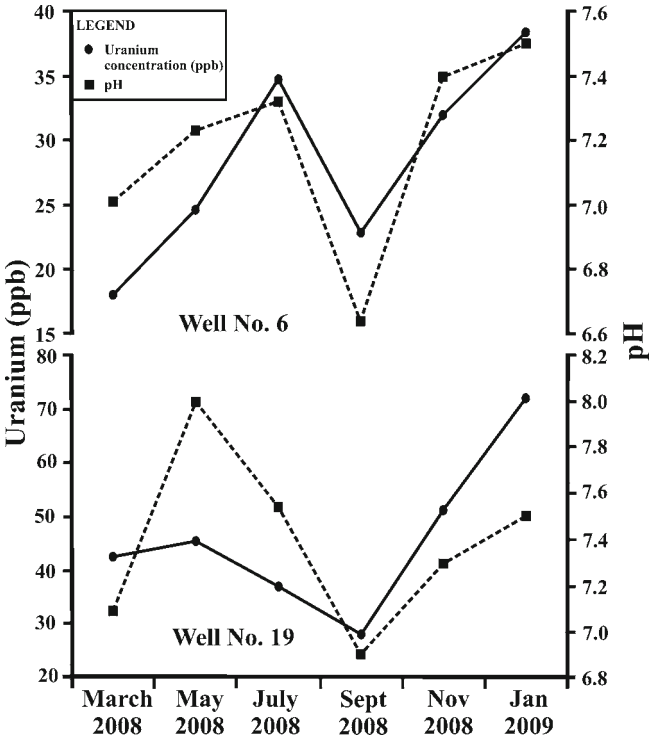

Figure 8. Temporal variation in uranium concentration (ppb) and pH.

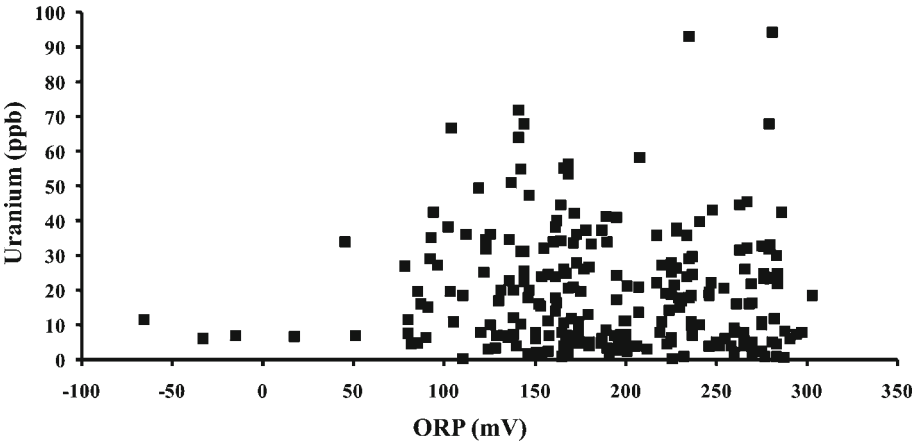

Figure 9. Relationship between uranium concentration (ppb) and ORP (mV).

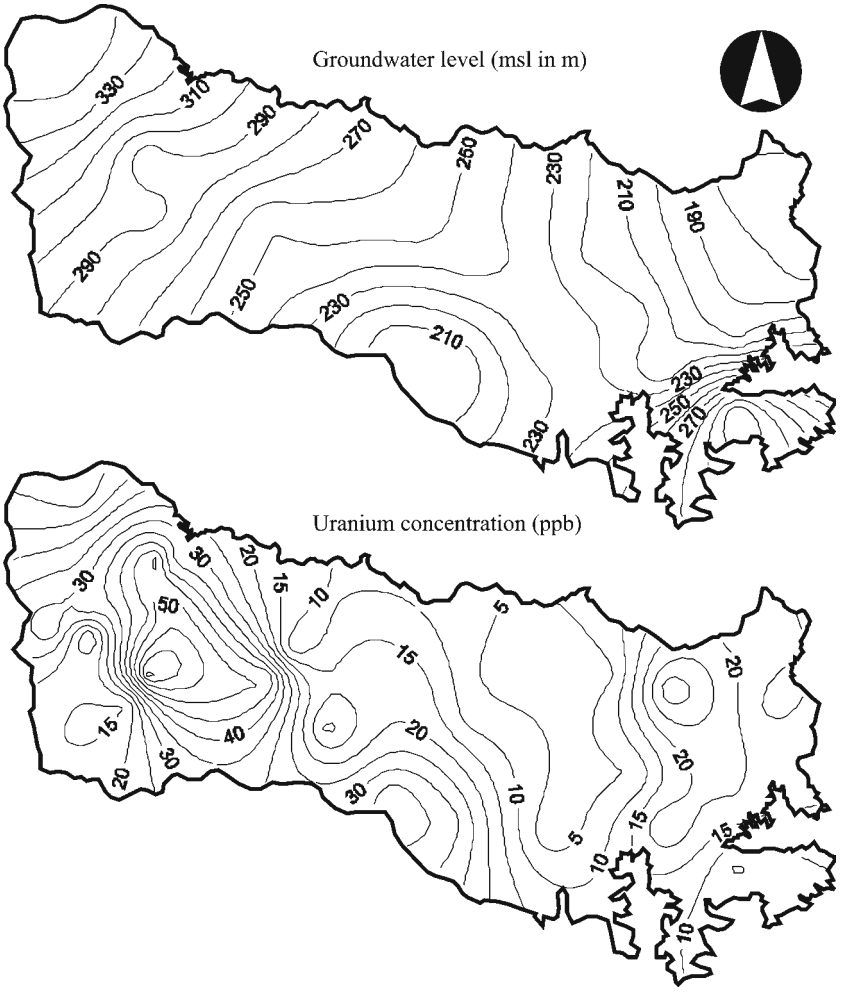

Figure 10. Regional groundwater level (msl in m) and uranium concentration (ppb) in November 2008.

is considered as the limit for this study. Of the total 236 groundwater samples collected and analyzed during the duration of this study, 21.6% of the groundwater samples had uranium above the USEPA limit. Uranium concentration generally is controlled by the changes in pH and ORP. In this area the uranium concentration in groundwater varies as in the case of pH (figure 8). Uranium concentration in groundwater has to be usually higher in oxidised condition which is evident from figure 9 where uranium concentration increases along with ORP.

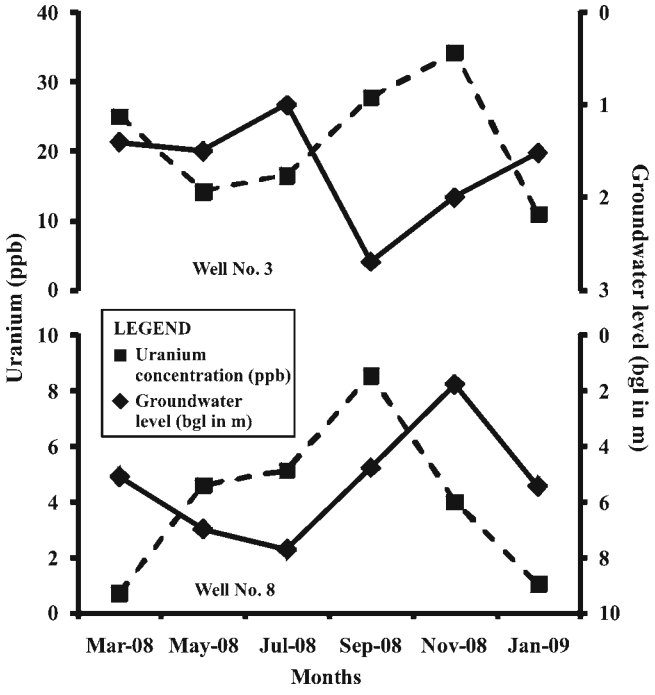

Figure 11. Temporal variation in groundwater level and uranium concentration.

Studying the spatial and temporal variation helps us to identify the contribution of various factors for the excess or low concentration of particular ions in groundwater. To understand the distribution of uranium in groundwater of the study area, iso concentration diagrams were prepared for all months.

Uranium concentration in groundwater of the study area varies spatially as well as temporally. Spatial variation in groundwater level and uranium concentration of the study area indicate that in general, the uranium concentration in groundwater increases along the flow direction (figure 10). The comparison between the uranium concentration and groundwater level in several wells indicate that the uranium concentration varies primarily due to recharge and discharge (figure 11). The rainfall recharge increases the groundwater level which in turn increases the uranium concentration in groundwater. The recharging water reacts with the weathered rocks in the unsaturated zone and the leached out uranium increases uranium concentration in groundwater. However, as the recharge continues concentration of uranium in groundwater begins to reduce due to dilution by comparatively fresh recharging water.

The granitic rocks which occur in most of the study area (figure 4) contain uranium in the range of 10.2–116 ppm (Shrivastava *et al* 1992). Interaction between these uranium rich weathered granitic rocks or top soil and groundwater has resulted in increase in uranium in this study area. The concentration of uranium is relatively high in three areas (figure 7). This is probably because of the presence of comparatively high uranium rich granitic rocks in these places.

In addition to the input derived from uranium rich rocks, use of phosphatic fertilisers for agricultural activity in this area may also add up to the uranium content in groundwater. Ioannides

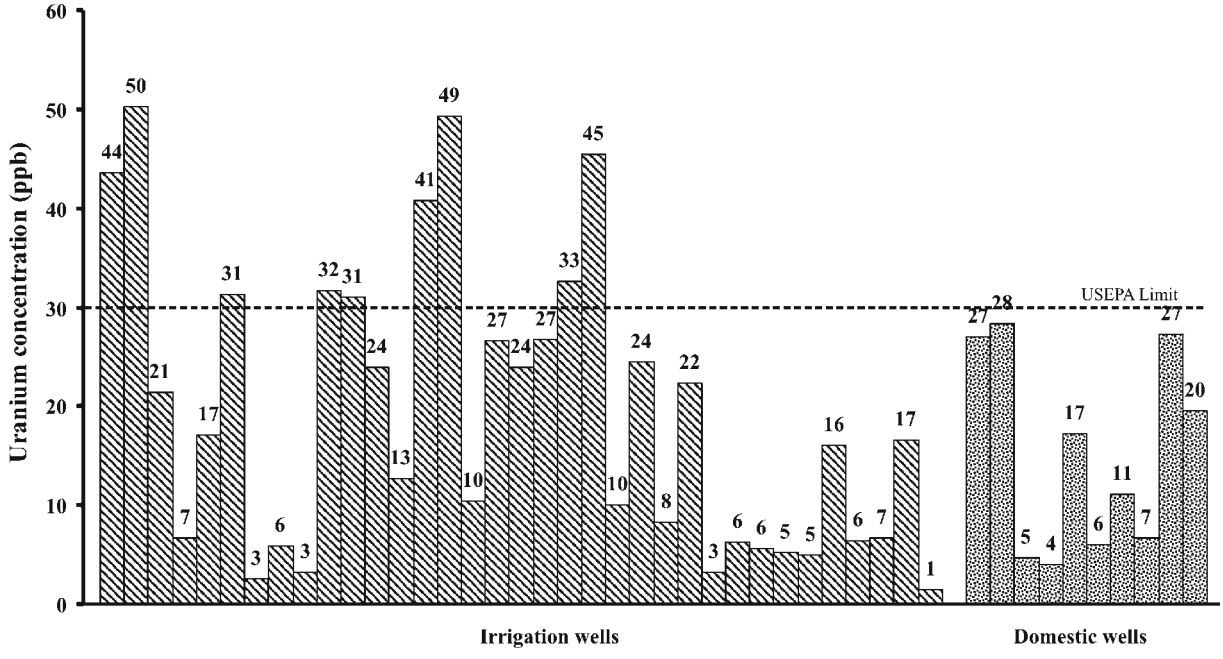

Figure 12. Average uranium concentration in groundwater (ppb) in irrigation and domestic wells.

Table 2. *Results of uranium concentration in fertilizers.*

Fertiliser	Uranium (μg/g)
Zinc sulphate	0.95
Potash	0.37
Ammonium sulphate	0.95
Urea	0.40
NPK complex	0.82

et al (1997); Azouazi *et al* (2001); Papastefanou *et al* (2006); Roselli *et al* (2009) have reported the natural radioactivity in phosphatic fertilisers. The range of uranium content in the phosphate rock is about 0.005–0.02%. These phosphate rocks serve as a source of phosphatic fertiliser to enhance the soil productivity in addition to other fertilisers. The total uranium resource in phosphate rock is estimated at 9×10^6 metric tonnes of uranium (Ragheb 2008). Hence, the phosphorous fertilisers manufactured form phosphate rocks may also contribute uranium to groundwater in the agricultural regions. Studies have shown that phosphate fertilisers possess uranium concentration ranging from 1 mg/kg to 68.5 mg/kg (Kawabata *et al* 2006). Concentration of fertilisers collected from this area varied from 0.37 ppb to 0.95 μg/g (table 2). On an average, about 128 kg/hectare of fertiliser is being used in this area (Directorate of Economics and Statistics 2010). Considering an average uranium concentration of 0.7 μg/g of fertiliser will lead to an addition of about 89,600 μg of uranium/year/hectare. Hence this also will add up to the uranium concentration in groundwater that resulted due to uranium rich rocks.

The average uranium concentration in groundwater of irrigation and domestic wells is given in figure 12. Although the uranium concentration in groundwater increases along the flow direction there is a difference between its concentration in domestic wells and irrigation wells. It is seen that more number of irrigation wells had uranium concentration above the permissible limit of 30 ppb. In case of domestic wells only one well had high uranium. Therefore the domestic wells have less concentration of uranium and thereby they can be considered to be safe for drinking purpose.

4. Conclusion

The uranium concentration in the groundwater of a part of Nalgonda district ranges from 0.2 to 68 ppb with an average of 18.5 ppb. About 21.6% of the groundwater samples had uranium concentration above the limit (30 ppb) set by USEPA for drinking purpose. The uranium content in groundwater varies primarily due to recharge and discharge which would have dissolved or leached the uranium from the weathered soil to the groundwater zone. The groundwater uranium concentration varied similar to the variation in pH. In general, the concentration of uranium in groundwater is high where the ORP is high. The uranium concentration was above 30 ppb in three areas during the entire period of study. In order to decrease the uranium concentration in groundwater in this area, artificial groundwater recharge techniques such as rainwater harvesting can be adopted. It is important to continuously monitor the groundwater quality in this area to study the impact of uranium mineralisation.

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