Applied Geochemistry 133 (2021) 105092

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Elevated uranium in drinking water sources in basement aquifers of southern India

Dan J. Lapworth [a,*], Bentje Brauns [b], Somsubhra Chattopadhyay [c,1], Daren C. Gooddy [a], Sian E. Loveless [a,2], Alan M. MacDonald [d], Andrew A. McKenzie [a], Sekhar Muddu [e], Siva N. V. Nara [f]

[a] British Geological Survey, Maclean Building, Crowmarsh Gifford, Wallingford, OX10 8BB, UK
[b] British Geological Survey, Environmental Science Centre, Keyworth, NG12 5GG, UK
[c] Ashoka Trust for Research in Ecology and the Environment, Bengaluru, Karnataka, 560064, India
[d] British Geological Survey, The Lyell Centre, Edinburgh, EH14 4AP, UK
[e] Department of Civil Engineering, Indian Institute of Science, Bengaluru, 560012, India
[f] Interdisciplinary Centre for Water Research (ICWaR), Indian Institute of Science, Bengaluru, Karnataka, 560012, India

ARTICLE INFO

Editorial handling by Huaming Guo

Keywords:
Uranium
Drinking waters
Speciation
Geogenic
Anthropogenic
India

ABSTRACT

Groundwater resources in the crystalline basement complex of India are crucial for supplying drinking water in both rural and urban settings. Groundwater depletion is recognised as a challenge across parts of India due to over-abstraction, but groundwater quality constraints are perhaps even more widespread and often overlooked at the local scale. Uranium contamination in basement aquifers has been reported in many parts of India, locally exceeding WHO drinking water guideline values of 30 μg/L and posing a potential health risk. In this study 130 water samples were collected across three crystalline basement catchments to assess hydrochemical, geological and anthropogenic controls on uranium mobility and occurrence in drinking water sources. Groundwaters with uranium concentrations exceeding 30 μg/L were found in all three study catchments (30% of samples overall), with concentrations up to 589 μg/L detected. There appears to be a geological control on the occurrence of uranium in groundwater with the granitic gneiss of the Halli and Bengaluru study areas having higher mean uranium concentrations (51 and 68 μg/L respectively) compared to the sheared gneiss of the Berambadi catchment (6.4 μg/L). Uranium – nitrate relationships indicate that fertiliser sources are not a major control on uranium occurrence in these case studies which include two catchments with a long legacy of intense agricultural land use. Geochemical modelling confirmed uranium speciation was dominated by uranyl carbonate species, particularly ternary complexes with calcium, consistent with uranium mobility being affected by redox controls and the presence of carbonates. Urban leakage in Bengaluru led to low pH and low bicarbonate groundwater hydrochemistry, reducing uranium mobility and altering uranium speciation. Since the majority of inhabitants in Karnataka depend on groundwater abstraction from basement aquifers for drinking water and domestic use, exposure to elevated uranium is a public health concern. Improved monitoring, understanding and treatment of high uranium drinking water sources in this region is essential to safeguard public health.

1. Introduction

India alone abstracts a third of the total annual global groundwater abstraction (Dalin et al., 2017). Groundwater depletion is a challenge across parts of India due to over abstraction (Wada et al., 2010), but groundwater quality constraints are perhaps even larger and are often overlooked at the local scale (MacDonald et al., 2016). Groundwater resources are crucial for supplying drinking water in both rural and urban settings across India due to the distributed nature of this large resource. However, co-occurrence of geogenic and anthropogenic contaminants constrain the availability of good quality drinking water in some regions (Lapworth et al., 2017; Coyte et al., 2019; Brindha et al.,

* Corresponding author.
E-mail address: djla@bgs.ac.uk (D.J. Lapworth).
[1] Current address: Department of Hydrology, Meteorology and Water Management, Warsaw University of Life Sciences, Poland.
[2] Current address: Environment Agency Crowmarsh Gifford, Wallingford OX10 8dBD

https://doi.org/10.1016/j.apgeochem.2021.105092
Received 16 April 2021; Received in revised form 10 September 2021; Accepted 11 September 2021
Available online 14 September 2021

2020). Geogenic sources of contamination can result in elevated arsenic (As) and fluoride (F) in groundwater throughout India (Podgorski et al., 2018; Mukherjee et al., 2020). This, together with the often limited groundwater treatment used in this region make the provision of good quality drinking water in many parts of India an ongoing challenge. Chemical drinking water quality is regularly monitored across India, although analysis for uranium (U) is still not routinely undertaken (CGWB 2020). There have been calls for improved monitoring and management of groundwater resources and improved public engagement to safeguard public health (Chakraborti et al., 2011; Francis et al., 2015; Coyte et al., 2018; CGWB 2020).

Uranium occurs naturally in the environment at variable concentrations in soils, rock and waters and can also be derived from anthropogenic activities such as mining and waste processing as well as agriculture (Schnug and Lottermoser, 2013). The hazardous chemical properties of U in drinking water can be a threat to human health. Research has indicated a link between elevated U in drinking water and chronic kidney disease (CKD) (e.g. Kurttio et al., 2002; Pinney et al., 2003; Scammell et al., 2019), although there are many environmental, lifestyle and genetic factors that lead to CKD (Jha et al., 2013). The World Health Organisation (WHO), and many other national regulators, have set a guideline value of 30 μg/L for U in drinking water (WHO, 2011), superseding an earlier provisional guideline value set at 15 μg/L (WHO, 2004). While India's Atomic Energy Regulatory Board has set a radiologically based limit of 60 μg/L for uranium in drinking water (AERB 2004), at present U is not included in India's national drinking water standards drawn up by the Bureau of India Standards. Groundwater sources with elevated U concentrations have been reported across India, both within the sedimentary systems of the Indo-Gangetic basin as well as within the basement complex which dominates central and southern India (Brindha and Elango, 2013; Coyte et al., 2018; CGWB 2020).

Uranium is closely associated with phosphate and iron minerals (e.g. Jerden et al., 2003; Scott et al., 2005). Elevated U concentrations in groundwater are in part controlled by the source rocks (e.g. granites) but also a range of other factors such as redox conditions, pH and the presence of carbonate ligands which form highly stable and mobile uranyl carbonate complexes. Uranium is most soluble as the uranyl ion in the +6 oxidation state $(UO_2)^{+2}$ and can be released from U-bearing minerals in aquifers, such as uraninite, and by dissolution and desorption processes with iron oxides, clay minerals and organic matter (Cumberland et al., 2016; Wazne et al., 2003). Hydrological processes such as recharge of oxidised groundwater and abstraction regimes can also potentially enhance U mobility and occurrence in groundwater (Alam and Cheng 2014; Lapworth et al., 2017, 2021; Erőss et al., 2018). Fertiliser use in agricultural areas has been proposed as a source of groundwater U in some regions (Schnug and Lottermoser 2013). Brindha et al. (2011) report that there may be up to 90 mg/ha/year of uranium added to land from the use of P based fertilisers in India, however, this is a relatively small source term compared to natural occurring concentrations found in granitic terrains (e.g. Langmuir 1978). Natural organic matter, e.g. humic acids, can also bind to and mobilise U under certain conditions (Bone et al., 2017). Recent studies have also shown that under reducing conditions oxidants such as nitrate (NO_3) can mobilise U by oxidizing U(IV) to U(VI), particularly in shallow aquifers where nitrate infiltration rates may exceed microbial reduction rates (e.g. Banning et al., 2013; Nolan and Weber, 2015).

The average upper crustal U content is 2.8 mg/kg (Taylor and McLennan 1985), with higher concentrations of 2–15 mg/kg in granite terrains (Langmuir 1978). The U content in granitic rocks is enhanced during magmatic differentiation (Rogers and Ragland, 1961). Patnaik et al. (2016) report 8–42 mg/kg from the Closepet granite (n = 6) and 415–1813 mg/kg from the Darshanapur granite in Peninsular India (n = 6). Rao et al. (1991) report U content of between 1.4 and 4 mg/kg for the Peninsular gneiss in India (n = 14). High (exceeding WHO guideline value of 30 mg/L) U groundwater concentrations have been reported in the basement complex of Southern India (e.g. Brindha and Elango, 2013). A small number of studies have previously reported elevated U within groundwaters in the Peninsular granitic gneiss complex of Karnataka. These are summarised in Table S1. Concentrations range from <detection limit to >2000 μg/L, with the highest values being reported in the Peninsular granitic gneiss of Kolar and Bengaluru (Babu et al., 2008; Mathews et al., 2015) and limestones in the Gogi and Gulbarga districts in northern Karnataka (Manoj et al., 2017; Kouser et al., 2019). Most studies have focussed on reporting the U occurrence in groundwater, association with particular lithologies and links to U prospecting. One study by Kouser et al. (2019) assessed hydrochemical controls, in terms of Eh-pH and uranyl carbonate complexes and uranium mineral saturation indices, for elevated U within the limestones of the Bhima group in northern Karnataka. Mathews et al. (2015) report high concentrations of U (up to 2027 μg/L) in groundwaters within the granitic aquifers in Bengaluru, with average concentrations of 92 μg/L and concentrations exceeding 30 μg/L in >60% of samples.

In this study, we compare U concentrations in drinking water sources in Karnataka, India, for groundwater, surface water and tap water samples, and investigate the geological and anthropogenic controls on U occurrence, mobility and speciation in groundwater, the main source of drinking water in this region. A comparison is made between observations from groundwaters in the Peninsular granitic gneiss (GGn) in Bengaluru and the surrounding area and those from the sheared gneiss (SGn) in the east of Karnataka in the Kabini catchment. We test the hypothesis that significantly lower U groundwater is present in the amphibole to granulite facies of the Dharwar Schists (supra-crustals) found in the Berambadi catchment (Meert et al., 2010; Valdiya and Sanwal 2017) compared to the granitic gneiss of the other two catchments with more widespread granite intrusions (Harris and Jayaram 1982). Anthropogenic influences from abstraction, urban leakage and fertiliser use on U occurrence are investigated through a comparison of urban and agriculturally intensive catchments within the granitic gneiss complex.

2. Methods

2.1. Study areas

The three study areas are all located on the Archean Gneiss Complex of Peninsular India characterised by tonalitic biotite gneisses (Harris and Jayaram, 1982). The K-rich granite-granodiorite-monzogranites of the Dharwar Craton are hybrid granitoids that formed through mixing between magmas derived from Na-rich tonalite-trondhjemite-granodiorites, sanukitoids, and K rich biotite- and mica granites (Laurent et al., 2014). The granitoids display large variations in concentration of trace elements, attributed to plagioclase accumulation or fluid-assisted mobilization of REEs (Ranjan et al., 2020). The Bengaluru study area is underlain by Precambrian migmatite, granodiorite, tonalite and gneiss, with local granitic intrusions (Fig. 1). Likewise, the geology of the Halli study area is granitic gneiss with local granitic and ultramafic intrusions (Srinivasan et al., 2015; KSRSTUC 1993). The Berambadi catchment is located on the Mysuru Plateau within the Kabini critical zone observatory (Sekhar et al., 2016) and is also underlain by granitic gneiss. In contrast to the other catchments the Berambadi is underlain by amphibolite-to granulite-facies metamorphic rocks, which have undergone local retrogressive metamorphism and shearing (Collins et al., 2020).

Hydrogeological investigations have been undertaken in all three areas and are described in detail elsewhere (Sekhar et al., 2016, 2017; Ballukraya and Srinivasan 2019; Collins et al., 2020) and are briefly summarised below. Groundwater levels in all three catchments are highly variable and are impacted by the geology and fracture network, seasonal monsoon recharge, abstraction both now and historically, and in the case of Bengaluru by urban leakage. Central Bengaluru has very shallow groundwater levels in some locations (*c.* 1 m below ground level

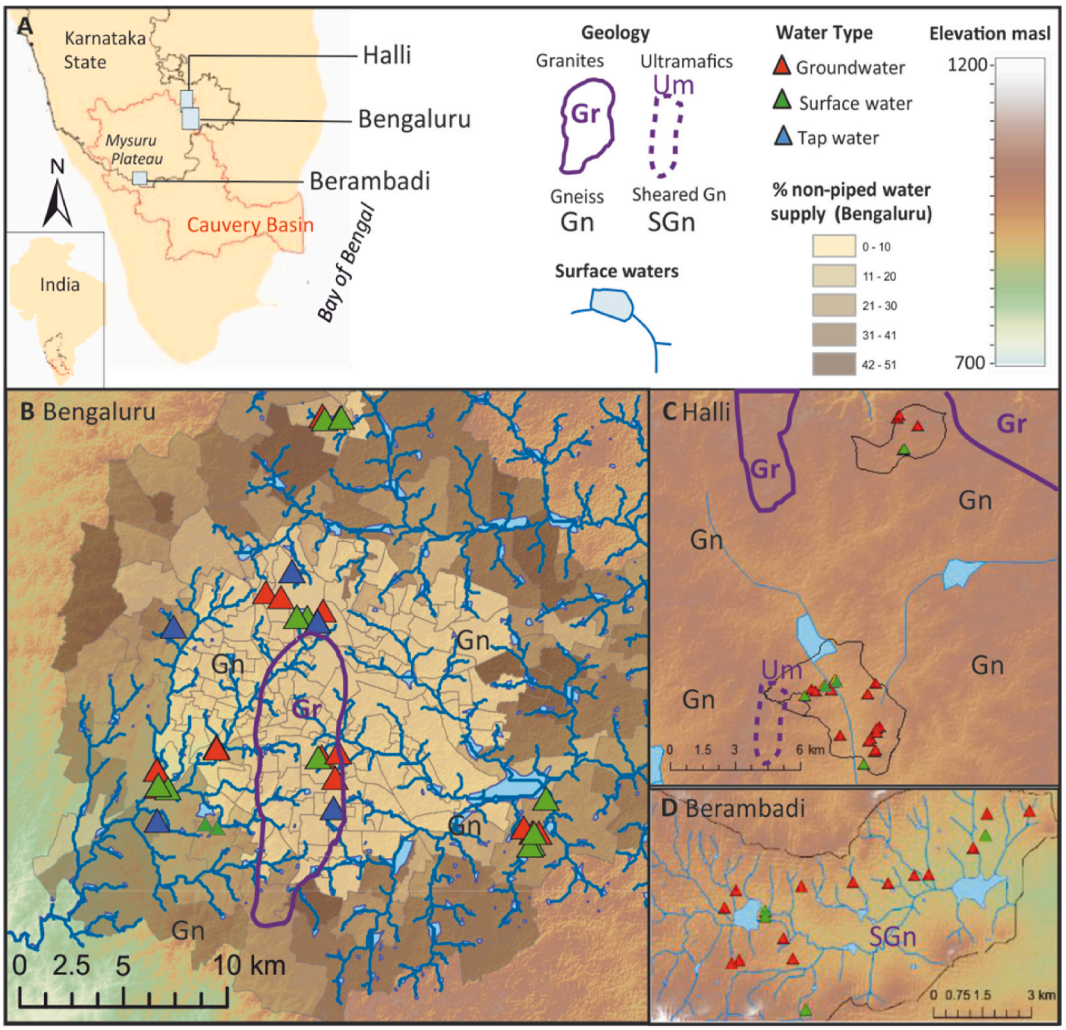

Fig. 1. Study area, sample sites and geology. A) Location of study catchments in relation to Karnataka and the Cauvery basin, B) Bengaluru study area, C) Halli study area, D) Berambadi study area. Data sources: geology information from KSRSTUC (1993), % non-piped water coverage in Bengaluru from Census of India (2012), elevation data from USGS SRTM data (http://earthexplorer.usgs.gov).

(mbgl)) but water tables are highly variable and are as deep as 70 meters at other locations (Sekhar et al., 2017). Within the Halli catchment, groundwater levels tend to be deeper than the other catchments (26–220 m bgl) and there is high spatial variability with highly contrasting levels observed at adjacent sites (Ballukraya and Srinivasan, 2019). Both the Bengaluru and Halli catchments have weathered zones that are typically 15–20 m thick, and are generally deeper than those found in the Berambadi catchment (Hedge and Subhash Chandra, 2014; Collins et al., 2020). In Berambadi, groundwater levels range from 10 to 70 mbgl, and a combination of increased abstraction for irrigation and variable monsoon recharge result in high temporal variability and an overall decline in groundwater tables between 2010 and 2018 across the catchment. This trend was rapidly reversed in 2018 which had a strong monsoon and subsequent recharge response in groundwater levels (Collins et al., 2020).

Fig. 1 shows the location of the three study areas sampled i) the city of Bengaluru (granitic gneiss), ii) the more rural area of the Thippagondanahalli catchment (Halli) *c.* 30 km north of Bengaluru (also on granitic gneiss) iii) the Berambadi catchment in the southern part of Karnataka (on sheared granitic gneiss). The catchments are mainly located in the Cauvery River Basin, with exception of the north-eastern part of Bengaluru, which is in the South Pennar basin. The Cauvery River Basin is India's third largest river basin which rises in the Western Ghats and drains into the Bay of Bengal (Fig. 1). Water samples were collected from a range of potential water sources, including groundwater, surface water and tap water. Surface water samples from rivers and lakes/tanks were collected for comparative purposes but are not typically used for domestic purposes unless alternatives are not available. Within all three catchments the surface drainage network connects a series of lakes and tanks which holds water up within the catchment.

2.2. Sampling and water analysis

A total of 130 samples were collected as part of this study during two sampling campaigns, one in October 2017 (post monsoon) and one in March–May 2018 (pre monsoon). Groundwater samples were collected from boreholes (n = 44) and a few open wells (n = 4) in all three catchments (sites n = 48, samples n = 92). Total borehole/well depths ranged from a few meters to 370 mbgl. Boreholes were purged using in-situ pumps prior to sampling and sampled once stable field readings for specific electrical conductance (SEC) and pH were obtained. Open wells were sampled using in-situ devices and consequently redox potential (Eh) and dissolved oxygen (DO) values were not obtained from these sites. Surface water sites (rivers and lakes/tanks) were sampled by taking grab samples in all three catchments (sites n = 18, samples n = 37). Tap water samples were also collected in Bengaluru for comparison with other waters (sites n = 5, samples n = 6).

For groundwater samples field measurements (using Mettler Toledo

sensors) of SEC, pH, Eh, dissolved oxygen and alkalinity (using a Hach titrator in duplicate) were taken, all 5 parameters were measured in 118 of the 130 samples collected. Eh, DO, pH measurements were taken in a flow-through cell to ensure a representative measurement of groundwater conditions was obtained. For surface water and tap water samples, SEC and pH measurements were undertaken in a bucket. All samples for anion and cation analysis were filtered in the field (0.45 μm filters) and stored in Nalgene containers prior to analysis. Cation samples were acidified with analytical grade nitric and hydrochloric acid. Field duplicates were taken for selected sites to assess reproducibility of results. A total of 72 groundwater samples for dissolved gas analysis for CFC-12 were taken, without atmospheric contact, in sealed containers using the USGS method (USGS 2020). These samples could only be taken from boreholes where there was no clear evidence of air intake in the rising main within the cased section of the borehole which could compromise the tracer analysis.

Cations, including U, were analysed by ICP-MS (Agilent 8900 Triple Quadrupole), anions by ion chromatography (Dionex ICS5000 dual line IC). CFC-12 was measured by gas chromatography (Agilent gas chromatographs) fitted with electron capture detectors (GC-ECD) with bespoke purge and trap system (e.g. Gooddy et al., 2006). All chemical analyses were undertaken at BGS geochemistry (accredited to ISO Standard 17025) and groundwater tracer laboratories in the UK. Anion and cation analysis was undertaken using UKAS accredited methods using NIST traceable standards and analysis checked with Certified Reference Materials.

To investigate U - dissolved organic carbon relationships in an urban setting, where pollution by organics is more likely, a subset of 24 sites from Bengaluru were analysed by fluorescence excitation emission matrix (EEM) spectroscopy to quantify dissolved organic matter and specifically to quantify relative amounts of humic acids in samples. Samples were analysed at the BGS, Wallingford (Oxfordshire) laboratory within three weeks of sample collection and samples were kept in the dark and filtered (0.45 micron silver filters) prior to storage in a refrigerator. Fluorescence EEMs were measured using a Cary Eclipse fluorescence spectrophotometer with a 1 cm path length. Scan settings were configured for emission from 280 to 500 at 2 nm intervals, and the excitation between 245 and 400 nm at 5 nm intervals. Milli-Q water was used as blank. Absorbance was blank corrected and measured in a 1 cm cuvette on a UV–vis spectrophotometer (Varian Cary 60) at 1 nm intervals from 800 to 200 nm. The EEMs for the samples were obtained by subtracting the blank EEM and undertaking absorbance and instruments corrections (Lakowicz, 2013) and were normalized to Raman units (RU) (Steadmon et al., 2003). See Lapworth et al. (2009) for further method details. Absorbance and instrument correction was applied to the fluorescence data and humic acid content was assessed using area averaging across humic regions of the EEM (Lapworth et al., 2009b).

We calculated the fluorescence index (FI) and beta to alpha (β:α) indices from EEMs, to better distinguish the sources and recently produced dissolved organic matter (McKnight et al., 2001; Wilson and Xenopoulos, 2009). FI was calculated as the ratio of fluorescence emission intensities at 470 and 520 nm with the excitation intensity of 370 nm (Cory and McKnight, 2005). β/α was calculated as the ratio of emission fluorescence intensity at 380 nm and the maximum emission fluorescence intensity observed between 420 and 435 nm at an excitation wavelength of 310 nm. All data processing was undertaken in R.

2.3. Geochemical modelling and statistical analysis

Mineral saturation indices (SI) for uraninite, calcite and iron oxyhydroxide and U speciation was calculated using the USGS PHREEQC software (Parkhurst and Appelo, 2013). The standard WATEQ4F database in PHREEQC (Ball and Nordstrom, 1991) was used and formation constants for ternary complexes of uranyl carbonate with Ca, Mg were added manually to PHREEQC using thermodynamic data from Richter et al. (2015). Redox potential (pE) was calculated using field Eh measurements. SI close to zero suggests solution equilibrium with the solid phase, SI $\ll 0$ indicates undersaturation where a particular mineral is unlikely to form. SI $\gg 0$ indicates supersaturation where the formation of a mineral is possible.

All statistical analyses were performed using R version 3.6.1 (The R Foundation). All plotting (boxplots, violin plots, cumulative frequency plots and jitter plots) were carried out using the ggplot2 package. Statistical tests were undertaken using nonparametric methods that did not assume normally distributed data (Spearman's rank correlation, Wilcoxon rank sum). Spatial plots were undertaken using ARCGIS®, version 10.3.

3. Results

3.1. Uranium concentrations in waters

Dissolved U concentrations for groundwaters, surface waters and tap waters are shown in Fig. 2. Only groundwater samples have concentrations in excess of the WHO guideline concentration of 30 μg/L. Summary statistics for U concentrations are presented in Table 1. The highest concentration was 589 μg/L within Bengaluru. All three catchments have groundwater concentrations that exceed 30 μg/L. Mean and standard errors for groundwater concentrations for Bengaluru, Halli and Berambadi were 68 (±30), 51 (±8.1) and 6.5 (±1.8) μg/L respectively. The proportion of groundwater samples with concentrations exceeding the WHO guideline value of 30 μg/L were 56%, 21% and 4% for Halli, Bengaluru and Berambadi catchments. The highest surface water U concentration is a river sample with 23 μg/L collected in the Vrishabhavathi River, downstream of a large wastewater treatment works (WTW) in Bengaluru. Uranium concentrations in the same river upstream of the WTW were 12 μg/L by comparison, suggesting that WTW are a potential source of U in rivers and lakes.

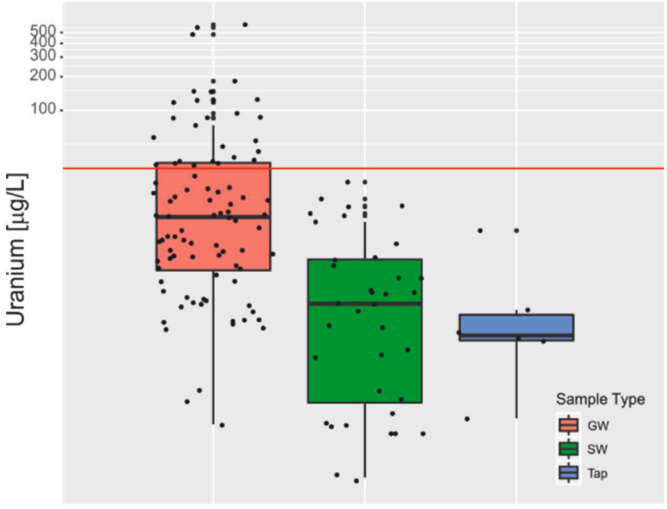

Fig. 2. Tukey box and jitter plot of U concentrations in groundwater, surface water and tap water samples. GW = groundwater (n = 92), SW = surface water (lakes and rivers, n = 37) and Tap = tap water from Bengaluru (n = 6). Horizontal red line shows the WHO drinking water guideline value of 30 μg/L. Note log (10) scale used on y axis. Whiskers for Fig. 2 and all subsequent box plots: lower whisker = smallest observation greater than or equal to lower hinge - 1.5 * IQR, upper whisker is greatest observation greater than or equal to upper hinge +1.5 * IQR. Outlier values are in line with the vertical whisker line, values to the left and right of the whisker show all individual data points (jitter plot), this is also the case for all subsequent box and jitter plots. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Uranium summary statistics for water sources and groundwater samples by catchment. All concentrations in µg/L.

Water type summary statistics			
Water type	Groundwater (n = 92)	Surface waters (n = 37)	Tap water (n = 6)
min	0.2	0.1	0.8
max	589	23	8.3
mean	43.7	4.3	2.3
median	11.1	1.8	1.1
%≥30 µg/L	29.7	0.0	0.0
Groundwater summary statistics			
Catchment	Halli (n = 36)	Bengaluru (n = 29)	Berambadi (n = 27)
min	4.7	0.2	1.1
max	183	589	47
mean	50.7	68.4	6.4
median	32.8	6.8	3.9
%≥30 µg/L	55.6	20.7	3.8

3.2. Groundwater nitrate concentrations

Groundwater nitrate concentrations were above the WHO guideline of 50 mg/L-NO_3 in a large proportion of sites, 36% overall, 80% of the groundwaters in Berambadi, 30% in Halli and 3% in Bengaluru. Surface water and tap water nitrate concentrations were below the WHO guideline value in all samples (See Supplementary Information).

3.3. Uranium concentrations and distributions in the three catchments

Fig. 3 shows graphical summaries of the distributions of U concentrations in groundwater for all three catchments. Fig. 3a shows a Tukey box-plot of results by catchment, Fig. 3b and c show the distributions by means of a violin and cumulative probability plots. Significantly higher median U concentrations were found in Halli catchment compared to the other two catchments (Bengaluru $p = 0.003$, Berambadi $p = 2.9^{-6}$, Wilcoxon rank sum test). It can be seen that the data from the Berambadi catchment has a unimodal distribution, in contrast to the distributions from the Halli and Bengaluru catchments which are both multimodal (see Fig. 3b and c). The Bengaluru catchment had one anomalously high site which had >480 µg/L on both sampling rounds. Fig. 3d shows a cross-plot of the two rounds of sampling, it can be seen that there is little difference between the two rounds for the majority of samples. However, the Halli catchment shows the most variability between sampling rounds with changes in both higher (n = 3) and lower (n = 3) concentrations between the two rounds. For 6 of the 7 samples with highest variability between rounds, concentrations were still found to be > 30 µg/L on both rounds. Overall, there was no significant difference between sampling rounds in the three catchments at the $p = 0.01$ level.

3.4. Relationship between uranium in groundwater and other parameters

Fig. 4 shows the relationships between U and other selected hydrochemical parameters in groundwaters. Highest U concentrations were found in groundwaters with alkalinity between 250 and 500 mg/L, pH between 6.5 and 7.5 and Eh > 200 mV (see Fig. 4a, c, 4d respectively), all waters had positive Eh values. Groundwater samples from Bengaluru had significantly lower median pH values compared to the other catchments (Halli $p = 1.1 \times 10^{-5}$, Berambadi $p = 7.1 \times 10^{-5}$, Wilcoxon test), with the highest U concentrations detected in samples with higher pH. Pooling results across all three catchments, significant positive correlations (Spearman rank) were found between groundwater U concentrations and molybdenum ($\rho = 0.76$, $p = 2.2 \times 10^{-16}$), TDS ($\rho = 0.51$, $p = 3.4^{-10}$), borehole depth ($\rho = 0.40$, $p = 6.9 \times 10^{-5}$) and

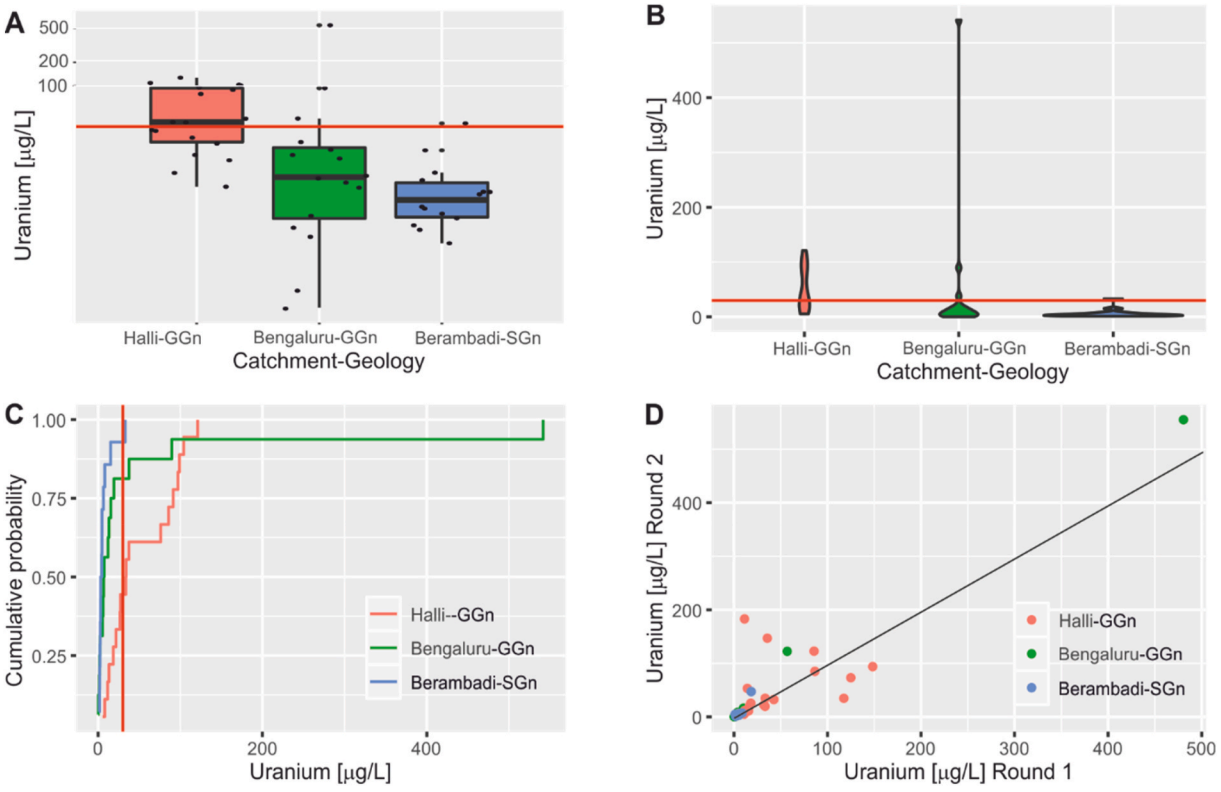

Fig. 3. Uranium distributions in groundwater survey catchments. A) Tukey box and jitter plot of groundwater U concentrations in the three study catchments, B) Violin plot of U concentrations, C) Cumulative frequency plots of U concentrations. D) Cross-plot of results from the repeat sampling from the two rounds (round 1 in October 2017 and round 2 in April 2018), black line shows the 1:1 line. Red horizontal and vertical lines show the U WHO drinking water guideline value of 30 µg/L. Log scale used on y axis (A) for clarity. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

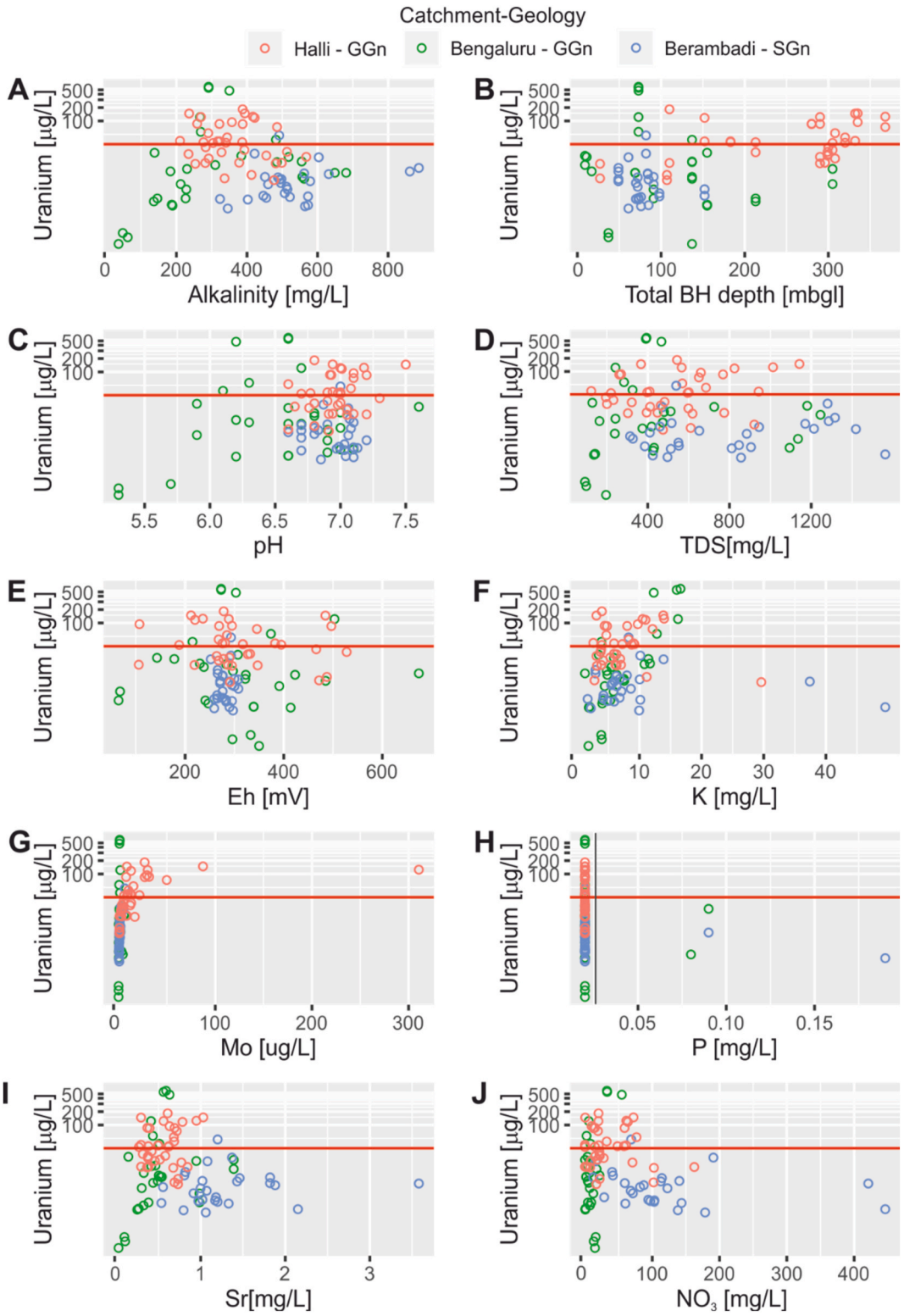

Fig. 4. Cross plots showing relationships between U (µg/L) and other hydrochemical and physical parameters. A) Alkalinity (mg/L), B) Total borehole depth (m below ground level) C) pH, D) TDS (mg/L) E) Field Eh (mV), F) Potassium (mg/L), G) Molybdenum (µg/L), and H) Total Phosphorus (mg/L), I) Strontium (mg/L), J) Nitrate (mg/L). Red horizontal and vertical lines show the U WHO drinking water guideline value of 30 µg/L. Black vertical line (H) shows the detection limit for total phosphorus, 0.025 mg/L. Note log scale used on y-axis. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

potassium ($\rho = 0.32$, $p = 0.001$) suggesting that water-rock interactions may be important in controlling U occurrence. However, no significant correlations were found between U and alkalinity, pH, Eh or Sr. There is a significant correlation between U and strontium for the Bengaluru catchment alone ($\rho = 0.56$, $p = 0.001$, Spearman rank) (Fig. 4i). If data for the two granitic gneiss catchments (Bengaluru and Halli) are pooled then correlations between U and potassium are still weak, but are higher ($\rho = 0.46$, $p = 0.0001$) than for all the data. Correlations between nitrate and U are also weak but significant for Bengaluru and Halli ($\rho = 0.34$, $p = 0.004$) but are not significant if data from Berambadi, which has significantly higher nitrate concentrations than the other catchments ($p = 2.1 \times 10^{-9}$, Wilcoxon test), are included.

A subset of samples from Bengaluru (n = 24) were used to explore relationships between dissolved organic matter content and U concentrations. There was a weak and insignificant correlation found between humic acid content and U concentrations ($\rho = 0.27$, $p = 0.18$, see Fig. S2a in supplementary material). However, there was a weak but significant (at the 0.05 level) correlation between humic acid content and alkalinity ($\rho = 0.44$, $p = 0.022$, see Fig. S2b in supplementary material) as might be expected due to the microbial breakdown of dissolved organic matter to form alkalinity in groundwater.

There is no clear relationship between CFC-12 concentrations and U concentrations (see Fig. S1 in supplementary material). However, it is noteworthy that all of the samples had detectable CFC-12 and for a significant proportion of samples CFC-12 concentrations exceeded concentrations expected for modern recharge (1.4 pmol/L), suggesting local sources of halon contamination leading to excess CFC-12 in groundwater are common.

3.5. Uranium speciation and mineral saturation indices

The results for calculated saturation indices and U speciation in groundwaters are summarised in Fig. 5 using Tukey boxplots and jitter plots. Uraninite is under saturated in all three catchments (Fig. 5a). Calcite and $Fe(OH)_3$ are saturated or supersaturated for the majority of samples in Halli and Berambadi but are undersaturated for a significant proportion of samples in Bengaluru (Fig. 5b and c). Uranium speciation for dominant species (as a % of total dissolved U) are shown in Fig. 5d–h. The dominant carbonate species was $Ca_2UO_2(CO_3)_3$ for most samples in all 3 catchments followed by $CaUO_2(CO_3)_3^{-2}$ and $MgUO_2(CO_3)_3^{-2}$ in

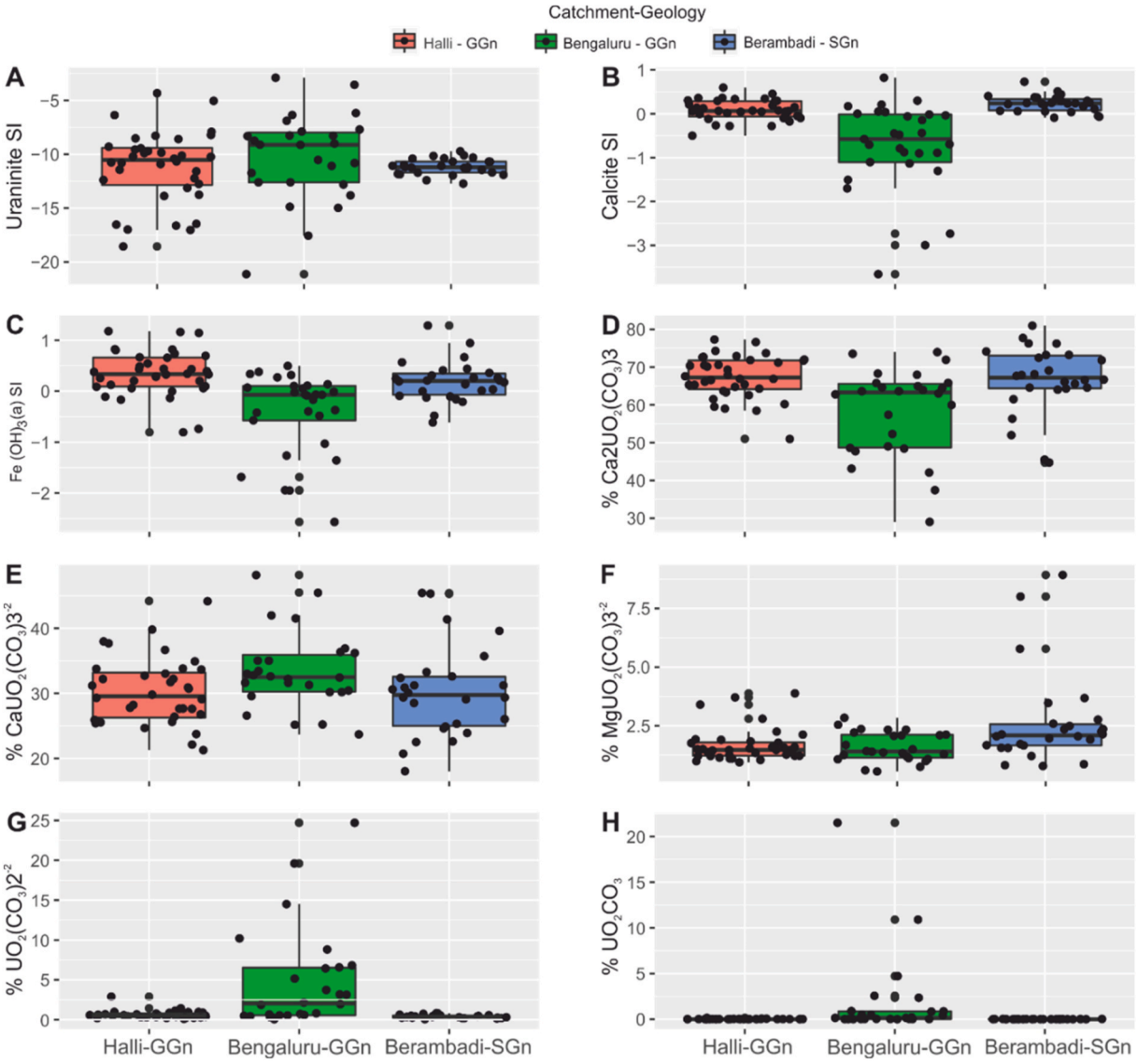

Fig. 5. Tukey box and jitter plots of selected groundwater mineral saturation indices and dominant U species (% of total dissolved U) found by study catchment. A) Uraninite saturation indices, B) Calcite saturation indices, C) Iron oxyhydroxide $Fe(OH)_3$ (a) saturation indices, D) $Ca_2UO_2(CO_3)_3$, E) $CaUO_2(CO_3)_3^{-2}$, F) $MgUO_2(CO_3)_3^{-2}$, G) $UO_2(CO_3)_2^{-2}$, H) UO_2CO_3.

the Halli and Berambadi catchments. In the Bengaluru samples there was a noticeable difference in speciation with $UO_2(CO_3)2^{-2}$ and UO_2CO_3 species more dominant than $MgUO_2(CO_3)_3^{-2}$ and accounting for up to 25% of dissolved U species (see Fig. 5g and h). Total phosphorus concentrations were low, below detection limit (0.025 mg/L) in all but four samples and therefore uranyl-phosphate species were not considered further. The four samples with detectable P all had U concentrations below 30 μg/L (Fig. 4j).

3.6. Spatial distribution of uranium in groundwater

Fig. 6 shows the spatial distribution of average U concentrations (from both rounds) in groundwater from the Bengaluru (Fig. 6a), Halli (Fig. 6b) and Berambadi (c) catchments in relation to catchment geology and mapped granite outcrops. It can be seen that the two sites in central Bengaluru with concentrations >30 mg/L, including the site with the highest U concentration, are associated with the large mapped granite intrusion in Bengaluru. However, there are sites in the north of the catchment with high U which are not in close proximity to mapped granite units (Fig. 6a). In contrast, there is no clear association between the mapped granite or ultramafic units in the Halli catchment and the high U groundwater sites found in this study (Fig. 6b).

4. Discussion

4.1. Uranium in drinking water and implications for health and exposure

Groundwater is the main source of drinking water and irrigation water across large parts of Peninsular India, as such the occurrence of high U groundwater concentrations is a potential concern for public health. This study has shown that surface water sources of drinking water in Bengaluru and within other catchments have significantly lower U concentrations than groundwater. However, treated piped tap water is not available in many settlements in Peninsular India and coverage in Bengaluru is not complete. River and tank water is still likely to pose a much greater risk to public health due to other risks associated

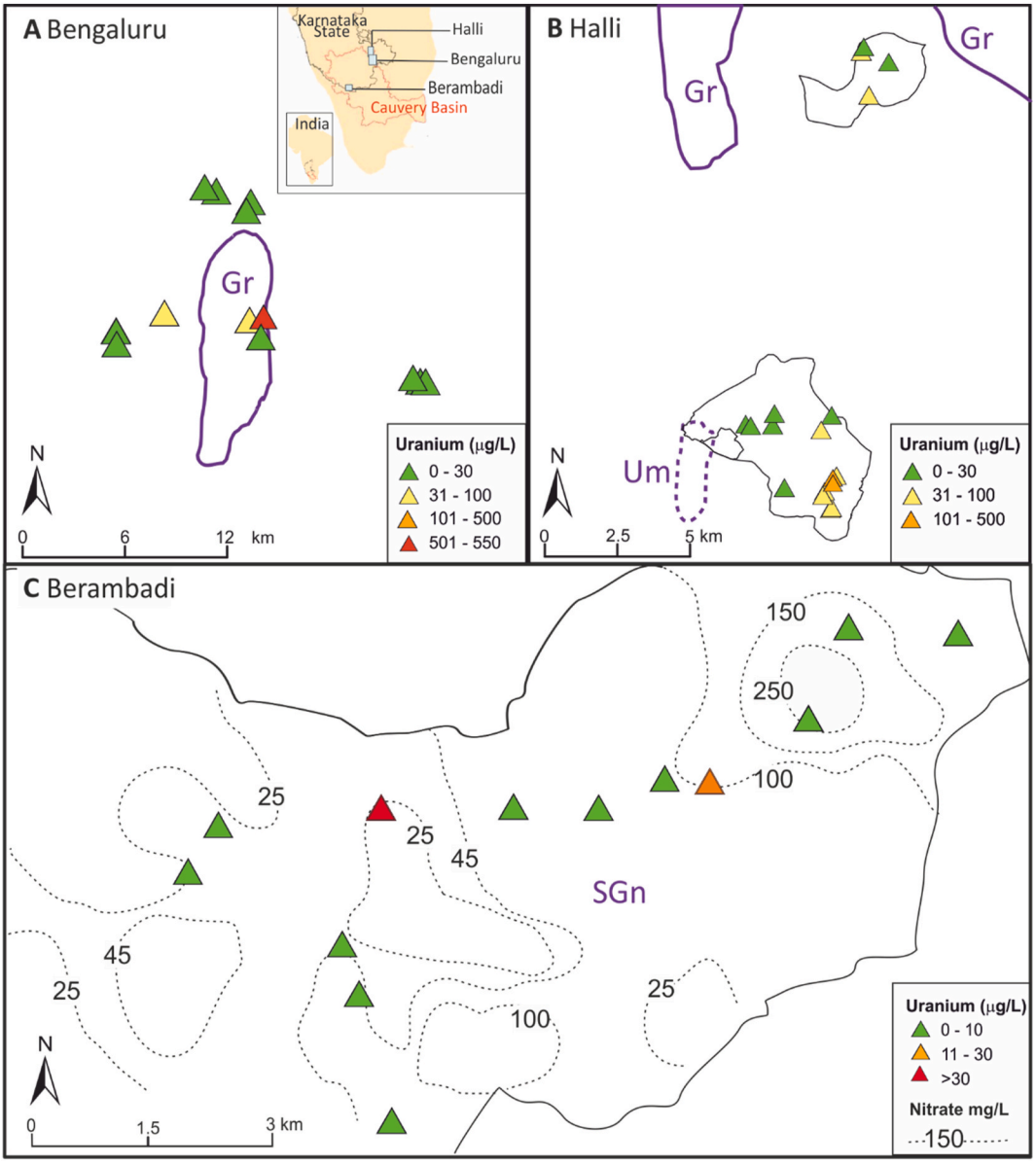

Fig. 6. Spatial distribution of mean groundwater U concentrations in the A) Bengaluru and B) Halli and C) Berambadi catchments. Mapped outcrops Gr = granites, Um = ultramafics, SGn = sheared granites. Contoured nitrate data (mg/L) sourced from Buvaneshwari et al. (2017). Solid black line shows the catchment boundary. For Berambadi the maximum average U concentration is 32.8 μg/L.

with other microbiological and chemical contaminants (Kumpel and Nelson 2013; Skariyachan et al., 2015).

Some groundwater U concentrations were found to exceed WHO guideline values of 30 μg/L in all three study catchments. However, the proportion of samples that exceeded 30 μg/L was considerably higher within the Halli and Bengaluru catchments (56% and 21% respectively) compared to the Berambadi catchment (4%).

The groundwater U concentrations found in this study are comparable with other recent studies within the granitic gneiss complex in Peninsular India (e.g. Babu et al., 2008, Brindha and Elango, 2013, Mathews et al., 2015, Manoj et al., 2017, Kouser et al., 2019). Groundwater in Bengaluru had a mean concentration of 68.4 μg/L, however, the median concentration was 6.8 μg/L showing the influence of a few high outlier values (Table 1). These distributions are comparable with results from previous groundwater studies in Bengaluru by Nagaiah et al. (2013) and Mathews et al. (2015) which reported maximum concentrations of 770 and 2027 μg/L, and median concentrations of 20.6 and 92.4 μg/L respectively. Uranium concentrations were found to vary greatly within short distances (Fig. 6a), this corroborates similar trends reported in Nagaiah et al. (2013), including the high U groundwater associated with the Bengaluru granite outcrop (Fig. 6a). The other high U concentrations found in Bengaluru and Halli are also possibly associated with granites which have either not been mapped or are not prominent at this scale. Only one published study from Chamarajanagar by Nagaraju et al. (2014) in the southwestern portion of the granitic gneiss in Karnataka has reported much lower concentrations, comparable with those found in this study in the Berambadi catchment. Both of these low U studies are in the same region and are located to the west of the large N–S trending Closepet granite emplacement in Karnataka (Moyen et al., 2001).

Maximum groundwater U concentrations found in this study in the Peninsular granitic gneiss are nearly twice those reported by Coyte et al. (2018) in the crystalline basement and alluvial lithologies in northern India, median concentrations are also higher than the alluvial lithologies in Coyte et al. (2018). The median concentration for the Halli catchment (32 μg/L) were comparable with the values reported for the Rajasthan crystalline basement by Coyte et al. (2018) and exceeded the WHO drinking water guideline value.

Uranium uptake via plants is also a possible additional source of dietary U where there is high U in soils and there is use of groundwater with high U for irrigation (Neves and Abreu 2009). Plant uptake is more likely if soil pH is acidic (Ebbs et al., 1998), which is the case in the western portion of the state which covers 39% of Karnataka (Wani et al., 2011). However, evidence from other studies suggests U exposure via plant consumption is not likely to be significant for human health outcomes even in conditions where U concentrations are high in soils and irrigation waters (Neves and Abreu 2009; Neves et al., 2012). Given the dependence on shallow groundwater in Peninsular India for drinking water the evidence from this and other studies from the Peninsular gneiss complex in Karnataka, suggest that a significant proportion of the population in this region may be exposed to chemically hazardous U concentrations via drinking water sources.

4.2. Geochemical controls on U occurrence, mobility and speciation

Oxidised groundwater conditions and the presence of bicarbonate are the most important factors in controlling the occurrence, mobility and speciation of U in groundwater (Elless and Lee 1998, Wazne et al., 2003; Smedley et al., 2006). Groundwater and soil pH are also directly linked to carbonate occurrence and speciation and thus also control U occurrence (Katsoyiannis 2007). These conditions are evident in the groundwater samples collected as part of this study and the presence of bicarbonate and *c*. neutral pH groundwaters (Fig. 4) are key factors in enhancing U mobility leading to elevated dissolved U in groundwater. This observation is consistent with speciation modelling conducted with PHREEQC, which predicted that uranyl carbonate species, especially ternary complexes with calcium ($Ca_2UO_2(CO_3)_3$ and $CaUO_2(CO_3)_3^{-2}$), are the predominant U complexes in groundwater. For a significant proportion of samples in Bengaluru the low pH, low bicarbonate and undersaturation with respect to calcite result in the higher predominance of $UO_2(CO_3)_2^{-2}$ and UO_2CO_3 and overall lower U concentrations (Fig. 3).

The hypothesis that groundwater within the amphibolite to granulite facies of gneiss of Peninsular India has lower U concentrations is supported by this study, however, in some cases high U can also be found within these zones – possibly related to less widespread granite intrusions. A geological source for the U in groundwater is inferred, this is corroborated by the correlation of U and K (Fig. 4f) likely due to mineral controls from biotite and feldspars within the host rocks (e.g. Idemitsu et al., 1994; Gao et al., 2017; Harris and Jayaram 1982). Results from the Berambadi catchment (Fig. 4) show the weakest relationship between K and U, ruling out potash fertilisers as a likely source of U in this catchment (Buvaneshwari et al., 2020). There were weak but statistically significant relationship observed between U and hydrochemical indicators of residence time or water-rock interactions (e.g. TDS – see Fig. 4) however no clear trends are present between U and other commonly used indicator of groundwater residence time such as Ca/Mg or Sr/Ca (see Supplementary Information Fig. S4). The equally suitable hydrochemical conditions for U mobility, but low U groundwaters found in the Berambadi suggests that the sheared granitic gneiss in this region may be depleted with respect to U compared to the other granitic gneiss catchments. Soil pH surveys by Wani et al. (2011) suggest that the predominantly acidic conditions in the western portion of Karnataka, where the Berambadi catchment is located, could lead to reduced U mobility in soils in this region, and is likely linked to changes in underlying geology. There is not enough evidence from these two studies alone to say if this is a regional trend, *i.e.* where generally lower groundwater U concentrations are found to the west of the Closepet granite in Karnataka, but this warrants further investigation.

4.3. Hydrological and anthropogenic factors that may influence U occurrence

Results from the groundwater residence time tracer CFC-12 show that these aquifer systems are all actively recharged and that this is occurring at considerable depths within the fractured gneiss system in all three catchments (Fig. S1). This builds on the results of Collins et al. (2020) from the Berambadi catchment and demonstrates that there is active recharge occurring across the Peninsular gneiss. The excess CFCs could be contamination of the subsurface from anthropogenic waste (e.g. Darling and Gooddy 2007), or possibly attributed to a regionally elevated atmospheric inputs (e.g. Ho et al., 1998).

Groundwaters in all three catchments are intensively abstracted for irrigation, and domestic and/or industrial use and this reflects the wider situation in southern India. Water tables in all three catchments show high seasonal variability and have been reported to be in significant decline in Berambadi and Halli, in part as a result of intensive abstraction (Sekhar et al., 2017; Tomer et al., 2020; Collins et al., 2020; Srinivasan et al., 2015). This has the effect of enhancing the oxidised conditions periodically within the aquifer system and potentially allowing these oxidised conditions to penetrate to greater depths than would be found under natural flow conditions enhancing the mobility and occurrence of trace elements including U (Ayotte et al., 2011; Lapworth et al., 2017). Intensive exploitation and highly variable groundwater levels is not unique to these case study areas, and is in fact a widespread phenomena across Karnataka and more widely within the basement aquifers of southern India which also have groundwaters where high U concentrations have been reported (e.g. Coyte et al., 2018; Dash et al., 2017).

Groundwater recharge from piped river water in Bengaluru is a significant component in the water budget of Bengaluru, particularly in central Bengaluru and is evidenced by piezometry, modelling and

through the use of hydrochemical tracers (Sekhar et al., 2017; Tomer et al., 2020). The impact of this low pH, low bicarbonate recharge source term is clearly seen in the hydrochemistry of a significant proportion of the Bengaluru samples (Fig. 4). This is likely to have a direct influence on reduced U mobility, lower occurrence and distinct U speciation and mineral saturation seen in some groundwaters beneath Bengaluru compared to the other catchments (Fig. 5). Conjunctive use of piped surface and groundwater is common in larger Indian cities and these processes may be important in several other large settlements within southern India and elsewhere.

There was no evidence from the subset of samples from Bengaluru that residual dissolved humic substances were a major control on U mobility in this catchment (see Fig. S2). However, there was a significant correlation between humic acid content and alkalinity highlighting the role of organic matter oxidation in the generation of dissolved inorganic carbon in groundwater. Regarding organic matter sources, fluorescence index values around 1.2 for tap waters indicate terrigenous origin, while values up to and exceeding 1.8 for some groundwaters and surface water indicate a mixture of terrigenous and microbial and autochthonous sources (Jaffé et al., 2008; McKnight et al., 2001) (see Fig. S3). Low values of β:α found in tap waters indicate older DOM, while values around 1 for several surface and groundwaters indicate a mixture of older and more fresh sources of organic matter (Wilson and Xenopoulos, 2009).

The lower nitrate concentrations observed in Bengaluru compared to Halli (Table 1, $p = 0.0002$, Wilcoxon rank sum) and the significantly lower median U concentrations compared to the groundwaters in the Halli catchment (Fig. 3, $p = 0.0003$, Wilcoxon rank sum) could be linked to the role of nitrate as an oxidising agent in more reduced zones of the groundwater system. The role of oxygen as an oxidising agent may be more limited in this setting due to the high water temperatures (mean groundwater temperature of 28 °C) and consequent lower oxygen solubility. Immobilisation of U(VI) is potentially constrained when oxidants such as oxygen and nitrate are recharged into more reducing zones within aquifer system (Banning et al., 2013; Wu et al., 2010).

Overall, there is no clear relationship between high U and high nitrate in our data sets (Fig. 4). The very high nitrate and low uranium groundwaters found in Berambadi rules out fertilisers as a dominant source of U within this case study area which is broadly representative of the geology and land use found in Karnataka and elsewhere in southern India (Fig. 6c). Furthermore, at the lower end of the Berambadi catchment, which has the longest history of irrigation and fertiliser use (Buvaneshwari et al., 2017, 2020; Collins et al., 2020), lower U concentrations are observed overall compared to the sampling points in the centre of the catchment (Fig. 6). Phosphorus groundwater concentrations were also found to be low in all three study areas (Fig. 4), likely due to P adsorption and immobilisation in the soil (e.g. Lair et al., 2009). This does not in rule out fertiliser U sources completely, but when put together with the evidence from groundwater nitrate concentrations (Fig. 4) does suggest that fertiliser sources are not the main controls for U occurrence even in intensively farmed catchments with high fertiliser use which have lower background geogenic U concentrations such as that found in Berambadi (Fig. 6).

These hydrological and anthropogenic factors, together with the geological controls on hydrochemistry and U mineralisation in the granitic gneiss provide conditions in which U is present, highly mobile and likely to occur in groundwater sources in concentrations exceeding WHO drinking water guideline values. Fertiliser U sources cannot be ruled out, but this study shows that they are not likely to be the dominant source of U in this hydrogeological setting. While groundwater abstraction alone is not the cause of high U groundwater in this region, it may enhance U mobility under the right conditions and could lead to the deterioration of deeper, previously lower U groundwater within the Peninsular gneiss complex through changes in redox conditions.

4.4. Global significance and implications

For high U to occur in groundwater there needs to be a significant source of U and suitable redox and hydrochemical conditions to mobilise and maintain the U in solution. Both these conditions are met in this study, with consequent elevated U, highlighting the potential hazard that granitic aquifers can pose globally to water supply. Intrusive granitic rocks have relatively high U content (Langmuir 1978) and are widely distributed, accounting for approximately 9% of exposed geology, while total shield rocks (intrusive igneous and metamorphic rocks) comprise around 34% (Suchet et al., 2003). Shield rocks underly the majority of Peninsular India, and account for around 30% of the exposed geology in India (Narasimhan 2006) and 34% in Africa (MacDonald and Calow., 2009). Oxic conditions within shallow granitic aquifers are common across many continents (e.g. Sidborn and Neretnieks, 2007; Lapworth et al., 2013; Coyte and Vengosh 2020; Adithya et al., 2019; Abiye and Bhattacharya 2019) and are linked to high U mobility in many cases. Furthermore, due to the low specific storage of granitic aquifers, which are commonly exploited for water supply, large variations in water levels are common as a result of either pumping and/or natural recharge processes (Chilton and Foster 1995; Reddy et al., 2009; Macdonald and Edmunds., 2014) which can potentially enhance oxic conditions suitable for U stability in solution (Ayraud et al., 2006; Tarits et al., 2006). This is particularly evident in wellfields for the growing towns and cities across Peninsular India and Africa which extract water from basement aquifers (e.g. Sekhar et al., 2017; Maréchal et al., 2018; Foster et al., 2012).

Evidence for the importance of anthropogenic sources of U, particularly from fertiliser sources, has been presented in many settings (Liesch et al., 2015; Lyons et al., 2020), including other parts of India (Brindha et al., 2011; Mathivanan et al., 2021), and often uses nitrate data to corroborate this hypothesis (e.g. Liesch et al., 2015). In this study, the use of nitrate and other hydrochemistry in oxic aquifers provides evidence that geological and hydrogeological controls are also clearly important in determining U occurrence in basement groundwater, and as such our findings are widely applicable to other regions. This is not only the case for granitic basement aquifers, but also apparent within sedimentary aquifers which are derived from weathering of shield rocks (Böhlke et al., 2007; Ayotte et al., 2011; Lapworth et al., 2017).

This study highlights the importance of low pH and low bicarbonate recharge of pipe leakage water from surface water sources in reducing U concentrations and modifying U speciation. Conjunctive use of surface water for urban water supply is common in India and elsewhere globally (Mukherjee et al., 2010; Foster et al., 2011), and leakage rates are often high (c.40%, Agrawal 2008; Kadu et al., 2015), as such this process may be occurring beneath other urban aquifers also, and may help limit U concentrations in some urban groundwater supplies.

The population of Southern India exceeds 250 million (Census 2011). There are hundreds of millions of inhabitants in India and elsewhere globally that are reliant on groundwater from shallow granitic aquifers (e.g. Hess et al., 1985; MacDonald et al., 2012; Mukherjee et al., 2015), some of which are potentially exposed to high concentrations of U in drinking water and water used for preparation of food (e.g. Orloff et al., 2004; Shin et al., 2016). 30% of samples in this study exceed WHO drinking water guideline values of 30 μg/L for U in drinking water. Links to cancer via U water sources have been highlighted in studies in USA and Sri Lanka (Wagner et al., 2011; Chandrajith et al., 2010). The WHO (2011) life average daily dose limit for U is 0.001 mg/kg/d, which would be exceeded in the majority of groundwater sites in this study if standard usage volumes were assumed. Compared to other hazardous naturally occurring elements, such as arsenic which are much more widely reported, the distribution of U in granitic aquifers remains poorly understood and warrants further investigation. Furthermore, treatment for U (or other contaminants) is rarely undertaken in many settings where naturally high U may be present and there are often limited alternatives

for water supply (e.g. across Asia and Africa) so understanding the occurrence in raw groundwater sources is essential to mitigate human health implications.

5. Conclusions

This study reports high U groundwaters from three catchments within the basement aquifers of southern India. Key conclusions are summarised below:

Groundwater U concentrations exceeding the WHO drinking water guideline value of 30 μg/L were found in all three study catchments (30% of sites overall), with concentrations up to 589 μg/L detected in this study. Higher U concentrations in groundwaters were found within the granitic gneiss (GGn) compared to the sheared granitic gneiss (SGn). Although concentrations can exceed 30 μg/L in both lithologies, there is a clear geological source that dominates and there are important hydrochemical controls (e.g. redox conditions, pH and carbonate species) on the occurrence and speciation of U in groundwater, with the Halli and Bengaluru (GGn) having higher maximum and mean U concentrations compared to the Berambadi catchment on the SGn. Paired U and nitrate observations from the three catchments support the hypothesis of a geological source rather than a fertiliser source of U.

Speciation modelling confirmed uranyl carbonate species, especially ternary complexes with calcium, dominate U speciation. Urban leakage from tap water in Bengaluru leads to a distinct low pH and low bicarbonate groundwater hydrochemistry, reducing U mobility and changing U speciation for a proportion of groundwaters in Bengaluru. Intense abstraction is important in sustaining oxidised groundwater conditions and may enhance U concentrations within these groundwaters compared to baseline conditions.

Uranium concentrations found in this study are comparable or exceed high concentrations found elsewhere in India and in other similar settings globally, in both the crystalline basement and alluvial aquifer systems. Evidence from this and other studies show that high U groundwater is pumped from the gneiss complex in Karnataka for drinking water and domestic use posing a potential risk to public health. Further work is needed to understand the public health risks posed by U in groundwater in granitic aquifers and improved monitoring and treatment of high U drinking water sources in this region is essential to safeguard public health.

Funding sources

Fieldwork was funded under the UPSCAPE project of the Newton-Bhabha programme “Sustaining Water Resources for Food, Energy and Ecosystem Services”, funded by the UK Natural Environment Research Council (NERC-UKRI) and the Indian Ministry of Earth Sciences (MoES), grant number(s) NE/N016270/1 and MoES/NERC/IA-SWR/P1/08/2016-PC-II. Interpretation and write up were funded by the British Geological Survey NC-ODA Grant NE/R000069/1: Geoscience for Sustainable Futures.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Amritha Thankachan, Renuka Prasad and Subhash Chandra are thanked for local guidance in Bengaluru, their valuable advice, and field support. Veena Srinivasan (ATREE) is thanked for overseeing ATREE activities, participating in discussions early on in the project on possible sampling sites and facilitating access to sites in the Halli catchment. We acknowledge GIS support provided by ATREE ecoinformatics lab. The Berambadi catchment is an Environmental Research Observatory (M-TROPICS https://mtropics.obs-mip.fr/itself part of OZCAR: The French Network of Critical Zone Observatories http://www.ozcar-ri.org/ozcar/). We thank all the private borehole and well owners for access to sites sampled as part of this study. We thank Carole Arrowsmith, NIGL — BGS Keyworth, for stable isotope analysis, and Michael Watts, Simon Chenery, Tom Barlow, Elliott Hamilton and Andy Marriott at BGS are thanked for providing the chemical analysis. British Geological Survey (BGS-UKRI) publish with the permission of the Director of BGS. The views and opinions expressed in this paper are those of the authors alone.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apgeochem.2021.105092.

References

Abiye, T.A., Bhattacharya, P., 2019. Arsenic concentration in groundwater: archetypal study from South Africa. Groundwater for Sustainable Development 9, 100246.
Adithya, V.S., Chidambaram, S., Keesari, T., Mohokar, H.V., Prasanna, M.V., 2019. Occurrence of uranium in groundwater along the lithological contacts in Central Tamilnadu, India: an isotope hydrogeochemical perspective. Exposure and Health 11 (4), 277–290.
Agrawal, P.C., 2008. Performance Improvement Planning: Designing an Effective Leakage Reduction and Management Program. World Bank Policy Research Working Paper, p. 44126.
Alam, M.S., Cheng, T., 2014. Uranium release from sediment to groundwater: influence of water chemistry and insights into release mechanisms. J. Contam. Hydrol. 164, 72–87.
Atomic Energy Regulatory Board (AERB), 2004. Drinking Water Specifications in India. Department of Atomic Energy, Mumbai, India.
Ayotte, J.D., Szabo, Z., Focazio, M.J., Eberts, S.M., 2011. Effects of human-induced alteration of groundwater flow on concentrations of naturally-occurring trace elements at water-supply wells. Appl. Geochem. 26 (5), 747–762.
Ayraud, V., Aquilina, L., Pauwels, H., Labasque, T., Pierson-Wickmann, A.C., Aquilina, A. M., Gallat, G., 2006. Physical, biogeochemical and isotopic processes related to heterogeneity of a shallow crystalline rock aquifer. Biogeochemistry 81 (3), 331–347.
Babu, M.N.S., Somashekar, R.K., Kumar, S.A., Shivanna, K., Krishnamurthy, V., Eappen, K.P., 2008. Concentration of uranium levels in groundwater. Int. J. Environ. Sci. Technol. 5, 263–266.
Ball, J.W., Nordstrom, D.K., 1991. WATEQ4F – User’s Manual with Revised Thermodynamic Data Base and Test Cases for Calculating Speciation of Major, Trace and Redox Elements in Natural Waters. U.S. Geological Survey Open-File, Report 90-129, p. 185.
Ballukraya, P.N., Srinivasan, V., 2019. Sharp variations in groundwater levels at the same location: a case study from a heavily overexploited, fractured rock aquifer system near Bengaluru, South India. Curr. Sci. 117 (1), 130–138.
Banning, A., Demmel, T., Rude, T.R., Wrobel, M., 2013. Groundwater uranium origin and fate control in a river valley aquifer. Environ. Sci. Technol. 47, 13941–13948.
Böhlke, J.K., Verstraeten, I.M., Kraemer, T.F., 2007. Effects of surface-water irrigation on sources, fluxes, and residence times of water, nitrate, and uranium in an alluvial aquifer. Appl. Geochem. 22 (1), 152–174.
Bone, S.E., Dynes, J.J., Cliff, J., Bargar, J.R., 2017. Uranium (IV) adsorption by natural organic matter in anoxic sediments. Proc. Natl. Acad. Sci. Unit. States Am. 114 (4), 711–716.
Brindha, K., Elango, L., 2013. Occurrence of uranium in groundwater of a shallow granitic aquifer and its suitability for domestic use in southern India. J. Radioanal. Nucl. Chem. 295 (1), 357–367.
Brindha, K., Elango, L., Nair, R.N., 2011. Spatial and temporal variation of uranium in a shallow weathered rock aquifer in southern India. J. Earth Syst. Sci. 120 (5), 911–920.
Brindha, K., Paul, R., Walter, J., Tan, M.L., Singh, M.K., 2020. Trace metals contamination in groundwater and implications on human health: comprehensive assessment using hydrogeochemical and geostatistical methods. Environ. Geochem. Health 42 (11), 3819–3839.
Buvaneshwari, S., Riotte, J., Sekhar, M., Kumar, M.M., Sharma, A.K., Duprey, J.L., Audry, S., Giriraja, P.R., Praveenkumarreddy, Y., Moger, H., Durand, P., 2017. Groundwater resource vulnerability and spatial variability of nitrate contamination: insights from high density tubewell monitoring in a hard rock aquifer. Sci. Total Environ. 579, 838–847.
Buvaneshwari, S., Riotte, J., Sekhar, M., Sharma, A.K., Helliwell, R., Kumar, M.M., Braun, J.J., Ruiz, L., 2020. Potash fertilizer promotes incipient salinization in groundwater irrigated semi-arid agriculture. Sci. Rep. 10 (1), 1–14.
Census of India 2011, 2012. House Listing and Housing Census, Government of India.
Central Groundwater Board (CGWB), 2020. Uranium Occurrence in Shallow Aquifers of India (last accessed Feb 2021). http://cgwb.gov.in/WQ/URANIUM_REPORT_2020.pdf.

Chakraborti, D., Das, B., Murrill, M.T., 2011. Examining India's groundwater quality management. Environ. Sci. Technol. 45 (1), 27–33. https://doi.org/10.1021/es101695d.
Chandrajith, R., Seneviratna, S., Wickramaarachchi, K., Attanayake, T., Aturaliya, T.N. C., Dissanayake, C.B., 2010. Natural radionuclides and trace elements in rice field soils in relation to fertilizer application: study of a chronic kidney disease area in Sri Lanka. Environ. Earth Sci. 60, 193– 20.
Chilton, P.J., Foster, S.S.D., 1995. Hydrogeological characterisation and water-supply potential of basement aquifers in tropical Africa. Hydrogeol. J. 3 (1), 36–49. https://doi.org/10.1007/s100400050061.
Collins, S.L., Loveless, S.E., Muddu, S., Buvaneshwari, S., Palamakumbura, R.N., Krabbendam, M., Lapworth, D.J., Jackson, C.R., Gooddy, D.C., Nara, S.N.V., Chattopadhyay, S., 2020. Groundwater connectivity of a sheared gneiss aquifer in the Cauvery River basin, India. Hydrogeol. J. 28, 1371–1388.
Cory, R.M., McKnight, D.M., 2005. Fluorescence spectroscopy reveals ubiquitous presence of oxidized and reduced quinones in dissolved organic matter. Environ. Sci. Technol. 39 (21), 8142–8149.
Coyte, R.M., Singh, A., Furst, K.E., Mitch, W.A., Vengosh, A., 2019. Co-occurrence of geogenic and anthropogenic contaminants in groundwater from Rajasthan, India. Sci. Total Environ. 688, 1216–1227.
Coyte, R.M., Jain, R.C., Srivastava, S.K., Sharma, K.C., Khalil, A., Ma, L., Vengosh, A., 2018. Large-scale uranium contamination of groundwater resources in India. Environ. Sci. Technol. Lett. 5 (6), 341–347.
Coyte, R.M., Vengosh, A., 2020. Factors controlling the risks of co-occurrence of the redox-sensitive elements of arsenic, chromium, vanadium, and uranium in groundwater from the eastern United States. Environ. Sci. Technol. 54 (7), 4367–4375.
Cumberland, S.A., Douglas, G., Grice, K., Moreau, J.W., 2016. Uranium mobility in organic matter-rich sediments: a review of geological and geochemical processes. Earth Sci. Rev. 159, 160–185.
Dalin, C., Wada, Y., Kastner, T., Puma, M.J., 2017. Groundwater depletion embedded in international food trade. Nature 543, 700–704.
Darling, W.G., Gooddy, D.C., 2007. Assessing the applicability of global CFC and SF_6 input functions to groundwater dating in Britain. Sci. Total Environ. 387, 353–362.
Dash, R.K., Ramanathan, A.L., Yadav, S.K., Kumar, M., Kuriakose, T., Gautam, Y.P., 2017. Uranium in groundwater in India: a review. Journal of Applied Geochemistry 19 (2), 138–144.
Ebbs, S.D., Brady, D.J., Kochian, L.V., 1998. Role of uranium speciation in the uptake and translocation of uranium by plants. J. Exp. Bot. 49 (324), 1183–1190.
Elless, M.P., Lee, S.Y., 1998. Uranium solubility of carbonate-rich uranium-contaminated soils. Water, air. Soil Pollut 107, 147–162.
Erőss, A., Csondor, K., Izsák, B., Vargha, M., Horváth, Á., Pándics, T., 2018. Uranium in groundwater–The importance of hydraulic regime and groundwater flow system's understanding. J. Environ. Radioact. 195, 90–96.
Foster, S.D., Hirata, R., Howard, K.W., 2011. Groundwater use in developing cities: policy issues arising from current trends. Hydrogeol. J. 19 (2), 271–274.
Foster, S., Tuinhof, A., Van Steenbergen, F., 2012. Managed groundwater development for water-supply security in Sub-Saharan Africa: investment priorities. WaterSA 38 (3), 359–366.
Francis, M.R., Nagarajan, G., Sarkar, R., Mohan, V.R., Kang, G., Balraj, V., 2015. Perception of drinking water safety and factors influencing acceptance and sustainability of a water quality intervention in rural southern India. BMC Publ. Health 15 (1), 731.
Gao, X., Bi, M., Shi, K., Chai, Z., Wu, W., 2017. Sorption characteristic of uranium (VI) ion onto K-feldspar. Appl. Radiat. Isot. 128, 311–317.
Gooddy, D.C., Darling, W.G., Abesser, C., Lapworth, D.J., 2006. Using chlorofluorocarbons (CFCs) and sulphur hexafluoride (SF_6) to characterise groundwater movement and residence time in a lowland Chalk catchment. J. Hydrol. 330 (1–2), 44–52.
Harris, N.B.W., Jayaram, S., 1982. Metamorphism of cordierite gneisses from the Bangalore region of the Indian Archean. Lithos 15 (2), 89–98.
Hedge, G.V., Subhash Chandra, K.C., 2014. Piezometric water-level condition in Bangalore city, Karnataka, India. Curr. Sci. 106 (2), 156–159.
Hess, C.T., Michel, J., Horton, T.R., Prichard, H.M., Coniglio, W.A., 1985. The occurrence of radioactivity in public water supplies in the United States. Health Phys. 48 (5), 553–586.
Ho, D.T., Schlosser, P., Smethie, W.M., Simpson, H.J., 1998. Variability in atmospheric chlorofluorocarbons (CCl_3F and CCl_2F_2) near a large urban area: implications for groundwater dating. Environ. Sci. Technol. 32, 2377–2382.
Idemitsu, K., Obata, K., Furuya, H., Inagaki, Y., 1994. Sorption behavior of uranium (VI) on a biotite mineral. MRS Online Proc. Libr. 353 (1), 981–988.
Jaffé, R., McKnight, D., Maie, N., Cory, R., McDowell, W., et al., 2008. Spatial and temporal variations in DOM composition in ecosystems: the importance of long-term monitoring of optical properties. J. Geophys. Res.: Biogeosciences 113 (G4).
Jerden Jr., J.L., Sinha, A.K., Zelazny, L., 2003. Natural immobilization of uranium by phosphate mineralization in an oxidizing saprolite–soil profile: chemical weathering of the Coles Hill uranium deposit, Virginia. Chem. Geol. 199 (1–2), 129–157.
Jha, V., Garcia-Garcia, G., Iseki, K., Li, Z., Naicker, S., Plattner, B., Saran, R., Wang, A.Y. M., Yang, C.W., 2013. Chronic kidney disease: global dimension and perspectives. Lancet 382 (9888), 260–272.
Kadu, M.S., Dighade, R.R., 2015. Infrastructure leakage index and challenges in water loss management in developing countries. In: World Environmental and Water Resources Congress, vol. 2015, pp. 1322–1331.
Katsoyiannis, I.A., 2007. Carbonate effects and pH-dependence of uranium sorption onto bacteriogenic iron oxides: kinetic and equilibrium studies. J. Hazard Mater. 139 (1), 31–37.
Karnataka State Remote Sensing Technology Utilisation Centre — KSRSTUC, 1993. Ground Water Prospect of Bangalore City & its Environs. July,1993, a report by Karnataka State Remote Sensing Technology Utilisation Centre, Bangalore, p. 22.
Kouser, L., Raghavendra, H.U., Rao, K.N., 2019. Studies on uranium hydrogeochemistry in kamalavathi river basin, Gulbarga and yadgir districts, Karnataka. J. Geol. Soc. India 94 (6), 605–610.
Kumpel, E., Nelson, K.L., 2013. Comparing microbial water quality in an intermittent and continuous piped water supply. Water Res. 47 (14), 5176–5188.
Kurttio, P., Auvinen, A., Salonen, L., Saha, H., Pekkanen, J., Makelainen, I., 2002. Renal effects of uranium in drinking water. Environ. Health Perspect. 110, 337–342.
Lair, G.J., Zehetner, F., Khan, Z.H., Gerzabek, M.H., 2009. Phosphorus sorption-desorption in alluvial soils of a young weathering sequence at the Danube River. Geoderma 149, 39–44.
Lakowicz, J.R., 2013. Principles of Fluorescence Spectroscopy, third ed. Springer Science & Business Media, p. 954.
Langmuir, D., 1978. Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits. Geochem. Cosmochim. Acta 42 (6), 547–569.
Lapworth, D.J., Gooddy, D.C., Allen, D., Old, G.H., 2009. Understanding groundwater, surface water, and hyporheic zone biogeochemical processes in a Chalk catchment using fluorescence properties of dissolved and colloidal organic matter. J. Geophys. Res.: Biogeosciences 114 (G3).
Lapworth, D.J., Kinniburgh, D., 2009. An R script for visualising and analysing fluorescence excitation–emission matrices (EEMs). Comput. Geosci. 35 (10), 2160–2163.
Lapworth, D.J., MacDonald, A.M., Tijani, M.N., Darling, W.G., Gooddy, D.C., Bonsor, H. C., Araguás-Araguás, L.J., 2013. Residence times of shallow groundwater in West Africa: implications for hydrogeology and resilience to future changes in climate. Hydrogeol. J. 21 (3), 673–686.
Lapworth, D.J., Krishan, G., MacDonald, A.M., Rao, M.S., 2017. Groundwater quality in the alluvial aquifer system of northwest India: new evidence of the extent of anthropogenic and geogenic contamination. Sci. Total Environ. 599, 1433–1444.
Lapworth, D.J., Dochartaigh, B.Ó., Nair, T., O'Keeffe, J., Krishan, G., MacDonald, A.M., Khan, M., Kelkar, N., Choudhary, S., Krishnaswamy, J., Jackson, C.R., 2021. Characterising groundwater-surface water connectivity in the lower Gandak catchment, a barrage regulated biodiversity hotspot in the mid-Gangetic basin. J. Hydrol. 594, 125923.
Laurent, O., Martin, H., Moyen, J.F., Doucelance, R., 2014. The diversity and evolution of late-Archean granitoids: evidence for the onset of "modern-style" plate tectonics between 3.0 and 2.5 Ga. Lithos 205, 208–235.
Liesch, T., Hinrichsen, S., Goldscheider, N., 2015. Uranium in groundwater—fertilizers versus geogenic sources. Sci. Total Environ. 536, 981–995.
Lyons, W.B., Gardner, C.B., Welch, S.A., Israel, S., 2020. Uranium in Ohio, USA surface waters: implications for a fertilizer source in waters draining agricultural lands. Sci. Rep. 10 (1), 1–6.
MacDonald, A.M., Bonsor, H.C., Ahmed, K.M., Burgess, W.G., Basharat, M., Calow, R.C., Dixit, A., Foster, S.S.D., Gopal, K., Lapworth, D.J., Lark, R.M., 2016. Groundwater quality and depletion in the Indo-Gangetic Basin mapped from in situ observations. Nat. Geosci. 9 (10), 762–766.
MacDonald, A.M, Bonsor, H.C, Dochartaigh, B. É. Ó, Taylor, R.G, 2012. Quantitative maps of groundwater resources in Africa. Environ. Res. Lett. 7 (2), 024009.
MacDonald, A.M., Calow, R.C., 2009. Developing groundwater for secure rural water supplies in Africa. Desalination 248 (1–3), 546–556.
Macdonald, D.M.J., Edmunds, W.M., 2014. Estimation of groundwater recharge in weathered basement aquifers, southern Zimbabwe; a geochemical approach. Appl. Geochem. 42, 86–100. https://doi.org/10.1016/j.apgeochem.2014.01.003.
Manoj, S., Thirumurugan, M., Elango, L., 2017. An integrated approach for assessment of groundwater quality in and around uranium mineralized zone, Gogi region, Karnataka, India. Arab. J. of Geosci. 10 (24), 557.
Mathews, G., Nagaiah, N., Karthik Kumar, M.B., Ambika, M.R., 2015. Radiological and chemical toxicity due to ingestion of uranium through drinking water in the environment of Bangalore, India. J. Radiol. Prot. Off. J. Soc. Radiol. Prot. 35, 447–455.
Maréchal, J.C., Selles, A., Dewandel, B., Boisson, A., Perrin, J., Ahmed, S., 2018. An observatory of groundwater in crystalline rock aquifers exposed to a changing environment: hyderabad, India. Vadose Zone J. 17 (1), 1–14.
Mathivanan, M., Sabarathinam, C., Viswanathan, P.M., Senapathi, V., Nadesan, D., Indrani, G.G., Malaimegu, G., Kumar, S.S., 2021. Mobilization and health risk assessment of fertilizer induced uranium in coastal groundwater. Environ. Res. 111791.
McKnight, D., Boyer, E., Weterhoff, P., Doran, P., Kulbe, T., et al., 2001. Spectrofluorometric characterization of dissolved organic matter for indication of precursor organic material and aromaticity. Limnol. Oceanogr. 46 (1), 38–48.
Meert, J.G., Pandit, M.K., Pradhan, V.R., Banks, J., Sirianni, R., Stroud, M., Newstead, B., Gifford, J., 2010. Precambrian crustal evolution of peninsular India: a 3.0 billion year odyssey. J. Asian Earth Sci. 39 (6), 483–515.
Moyen, J.F., Martin, H., Jayananda, M., 2001. Multi-element geochemical modelling of crust–mantle interactions during late-Archaean crustal growth: the Closepet granite (South India). Precambrian Res. 112 (1–2), 87–105.
Mukherjee, A., Saha, D., Harvey, C.F., Taylor, R.G., Ahmed, K.M., Bhanja, S.N., 2015. Groundwater systems of the Indian sub-continent. J. Hydrol.: Reg. Stud. 4, 1–14.
Mukherjee, A., Sarkar, S., Chakraborty, M., Duttagupta, S., Bhattacharya, A., Saha, D., Bhattacharya, P., Mitra, A., Gupta, S., 2020. Occurrence, predictors and hazards of elevated groundwater arsenic across India through field observations and regional-scale odelling. Sci. Total Environ. 143511.

Mukherjee, S., Shah, Z., Kumar, M.D., 2010. Sustaining urban water supplies in India: increasing role of large reservoirs. Water Resour. Manag. 24 (10), 2035–2055.
Nagaiah, N., Mathews, G., Balakrishna, K.K., Rajanna, A.M., Naregundi, K., 2013. Influence of physico-chemical parameters on the distribution of uranium in the ground water of Bangalore, India. Radiat. Protect. Environ. 36 (4), 175–180.
Nagaraju, K.M., Chandrashekara, M.S., Pruthvi Rani, K.S., Paramesh, L., 2014. Estimation of radiation dose due to uranium in water to the public in chamarajanagar district, Karnataka state, India. Am. Int. J. Res. Sci. Technol. Eng. Math. 2, 144–147.
Narasimhan, T.N., 2006. Ground water in hard-rock areas of peninsular India: challenges of utilization. Ground Water 44 (2), 130–133.
Neves, O., Abreu, M.M., 2009. Are uranium-contaminated soil and irrigation water a risk for human vegetables consumers? A study case with Solanum tuberosum L., Phaseolus vulgaris L. and Lactuca sativa L. Ecotoxicology 18 (8), 1130–1136.
Neves, M.O., Abreu, M.M., Figueiredo, V., 2012. Uranium in vegetable foodstuffs: should residents near the Cunha Baixa uranium mine site (Central Northern Portugal) be concerned? Environ. Geochem. Health 34 (2), 181–189.
Nolan, J., Weber, K.A., 2015. Natural uranium contamination in major U.S. Aquifers linked to nitrate. Environ. Sci. Technol. Lett. 2, 215–220. https://doi.org/10.1021/acs.estlett.5b00174.
Orloff, K.G., Mistry, K., Charp, P., Metcalf, S., Marino, R., Shelly, T., Melaro, E., Donohoe, A.M., Jones, R.L., 2004. Human exposure to uranium in groundwater. Environ. Res. 94 (3), 319–326.
Parkhurst, D.L., Appelo, C.A.J., 2013. Description of Input and Examples for PHREEQC Version 3: a Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations (No. 6-A43). US Geological Survey.
Patnaik, S., Hegde, G.N., Panneerselvam, A., Verma, M.B., Mohanty, R., Rai, A.K., 2016. Geochemical behaviour of LREE, Y and Zr in uranium mineralized and non-mineralized granite from Darshanapur area, in the Gogi-Kurlagere fault zone, Bhima basin, Yadgiri district, Karnataka. J. Geol. Soc. India 88 (2), 151–158.
Pinney, S.M., Freyberg, R.W., Levine, G.H., Brannen, D.E., Mark, L.S., Nasuta, J.M., Tebbe, C.D., Buckholz, J.M., Wones, R., 2003. Health effects in community residents near a uranium plant at Fernald, Ohio, USA. Int. J. Occup. Med. Environ. Health 16 (2), 139–153.
Podgorski, J.E., Labhasetwar, P., Saha, D., Berg, M., 2018. Prediction odelling and mapping of groundwater fluoride contamination throughout India. Environ. Sci. Technol. 52 (17), 9889–9898.
Ranjan, S., Upadhyay, D., Abhinay, K., Srikantappa, C., 2020. Paleoarchean and neoarchean tonalite–trondhjemite–granodiorite (TTG) and granite magmatism in the western dharwar Craton, southern India: implications for archean continental growth and geodynamics. Precambrian Res. 340, 105630.
Rao, Y.B., Naha, K., Srinivasan, R., Gopalan, K., 1991. Geology, geochemistry and geochronology of the Archaean Peninsular gneiss around Gorur, Hassan district, Karnataka, India. Proc. Indian Acad. Sci. Earth Planet Sci. 100 (4), 399–412.
Reddy, D.V., Nagabhushanam, P., Sukhija, B.S., Reddy, A.G.S., 2009. Understanding hydrological processes in a highly stressed granitic aquifer in southern India. Hydrol. Process.: Int. J. 23 (9), 1282–1294.
Richter, A., Bok, F., Brendler, V., 2015. Data compilation and evaluation of U (IV) and U (VI) for thermodynamic reference database THEREDA (No. HZDR—065). Helmholtz-Zentrum Dresden-Rossendorf eV. Accessed Dec 2020. https://inis.iaea.org/collection/NCLCollectionStore/_Public/47/081/47081518.pdf?r=1&r=1, 61.
Rogers, J.J., Ragland, P.C., 1961. Variation of thorium and uranium in selected granitic rocks. Geochem. Cosmochim. Acta 25 (2), 99–109.
Scammell, M.K., Sennett, C.M., Petropoulos, Z.E., Kamal, J., Kaufman, J.S., 2019. Environmental and occupational exposures in kidney disease. Semin. Nephrol. 39 (3), 230–243.
Schnug, E., Lottermoser, B.G., 2013. Fertilizer-derived uranium and its threat to human health. Environ. Sci. Technol. 47, 2433–2434.
Scott, T.B., Allen, G.C., Heard, P.J., Randell, M.G., 2005. Reduction of U (VI) to U (IV) on the surface of magnetite. Geochem. Cosmochim. Acta 69 (24), 5639–5646.
Sekhar, M., Riotte, J., Ruiz, L., Jouquet, P., Braun, J.J., 2016. Influences of climate and agriculture on water and biogeochemical cycles: Kabini critical zone observatory. Proc. Indian Natl. Sci. Acad 82 (3), 833–846.
Sekhar, M., Tomer, S.K., Thiyaku, S., Giriraj, P., Murthy, S., Mehta, V.K., 2017. Groundwater level dynamics in Bengaluru city, India. Sustainability 10 (1), 26. https://doi.org/10.3390/su10010026.
Shin, W., Oh, J., Choung, S., Cho, B.W., Lee, K.S., Yun, U., Woo, N.C., Kim, H.K., 2016. Distribution and potential health risk of groundwater uranium in Korea. Chemosphere 163, 108–115.
Sidborn, M., Neretnieks, I., 2007. Long term redox evolution in granitic rocks: modelling the redox front propagation in the rock matrix. Appl. Geochem. 22 (11), 2381–2396.
Skariyachan, S., Mahajanakatti, A.B., Grandhi, N.J., Prasanna, A., Sen, B., Sharma, N., Vasist, K.S., Narayanappa, R., 2015. Environmental monitoring of bacterial contamination and antibiotic resistance patterns of the fecal coliforms isolated from Cauvery River, a major drinking water source in Karnataka, India. Environ. Monit. Assess. 187 (5), 279.
Smedley, P.L, Smith, B, Abesser, C, Lapworth, D.J, 2006. Uranium occurrence and behavior in British groundwater. CR/06/050/N British Geological Survey.
Srinivasan, V., Thompson, S., Madhyastha, K., Penny, G., Jeremiah, K., Lele, S., 2015. Why is the Arkavathy River drying? A multiple-hypothesis approach in a data-scarce region. Hydrol. Earth Syst. Sci. 19 (4), 1905–1917.
Steadmon, C, Markager, S, Bro, R, 2003. Tracing dissolved organic matter in aquatic environments using a new approach to fluorescence spectroscopy. Mar. Chem. 82, 239–254.
Suchet, A.P., Probst, J.L., Ludwig, W., 2003. Worldwide distribution of continental rock lithology: implications for the atmospheric/soil CO2 uptake by continental weathering and alkalinity river transport to the oceans. Global Biogeochem. Cycles 17 (2).
Tarits, C., Aquilina, L., Ayraud, V., Pauwels, H., Davy, P., Touchard, F., Bour, O., 2006. Oxido-reduction sequence related to flux variations of groundwater from a fractured basement aquifer (Ploemeur area, France). Appl. Geochem. 21 (1), 29–47.
Taylor, S.R., McLennan, S.M., 1985. The Continental Crust: its Composition and Evolution. Wiley-Blackwell, Oxford, p. pp328.
Tomer, S.K., Sekhar, M., Balakrishnan, K., Malghan, D., Thiyaku, S., Gautam, M., Mehta, V.K., 2020. A model-based estimate of the groundwater budget and associated uncertainties in Bengaluru, India. Urban Water J. https://doi.org/10.1080/1573062X.2020.1836237.
USGS, 2020. USGE Groundwater CFC Sampling Method. Last accessed in Dec 2020. https://water.usgs.gov/lab/chlorofluorocarbons/sampling/bottles/.
Valdiya, K.S., Sanwal, J., 2017. Mysore plateau and nilgiri massif. In: Valdiya, K.S., Sanwal, J. (Eds.), Developments in Earth Surface Processes. Elsevier, Amsterdam, the Netherlands, pp. 279–306.
Wada, Y., van Beek, L.P.H., van Kempen, C.M., Reckman, J.W.T.M., Vasak, S., Bierkens, M.F.P., 2010. Global depletion of groundwater resources. Geophys. Res. Lett. 37 (20), L20402. https://doi.org/10.1029/2010GL044571.
Wagner, S.E., Burch, J.B., Bottai, M., Puett, R., Porter, D., Bolick-Aldrich, S., Temples, T., Wilkerson, R.C., Vena, J.E., Hébert, J.R., 2011. Groundwater uranium and cancer incidence in South Carolina. Canc. Causes Contr. 22 (1), 41–50.
Wani, S.P., Sahrawat, K.L., Sarvesh, K.V., Baburao, M., Krishnappa, K., 2011. Soil Fertility Atlas for Karnataka, India. International Crops Research Institute for the Semi-Arid Tropics, p. 312.
Wazne, M., Korfiatis, G.P., Meng, X., 2003. Carbonate effects on hexavalent uranium adsorption by iron oxyhydroxide. Environ. Sci. Technol. 37, 3619–3624.
WHO, 2004. Guidelines for drinking-water quality, 3rd. World Health Organization, Geneva.
WHO, 2011. Guidelines for drinking-water quality, 4th. World Health Organization, Geneva.
Wilson, H.F., Xenopoulos, M.A., 2009. Effects of agricultural land use on the composition of fluvial dissolved organic matter. Nat. Geosci. 2 (1), 37–41.
Wu, W.M., Carley, J., Green, S.J., Luo, J., Kelly, S.D., Van Nostrand, J., Lowe, K., Mehlhorn, T., Carroll, S., Boonchayanant, B., Lofller, F.E., Watson, D., Kemner, K.M., Zhou, J.Z., Kitanidis, P.K., Kostka, J.E., Jardine, P.M., Criddle, C.S., 2010. Effects of nitrate on the stability of uranium in a bioreduced region of the subsurface. Environ. Sci. Technol. 44 (13), 5104–5111, 2010.