Hydrogeochemical processes governing Uranium mobility: Inferences from the anthropogenically disturbed, semi-arid region of India

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Abstract

Khetri Copper Belt, Rajasthan is anthropogenically active and geologically belongs to the Delhi supergroup. To study the active geochemical processes controlling the elemental mobility, representative groundwater samples were analysed for heavy metals and radionuclide (U). A maximum U concentration (average 87 $\mu$g L\textsuperscript{-1}) is observed in the quartzite mine zone whereas minimum (average 13 $\mu$g L\textsuperscript{-1}) is found to be in the copper mine zone samples. Despite the presence of Jaspura and Gothra granitoid in the copper mine zone, the abundance of U is low suggesting the scavenging of U by sulphides or iron oxides. Additionally, at the confluence of two geological groups, Fe concentration is found high with a low concentration of U which further confirms scavenging of U. In the region, aquifers are shallow which supports the weathering of minerals. Thus, a high concentration of U in groundwater is attributed to mineral dissolution, faults or fractures. It is evident from the results that in the absence of iron-bearing sulphides, U concentration in groundwater would be very high compared to the current concentration. It also indicates low concentration of U in the copper mine zone, area is covered with abundant quantity of Fe sulphide rich waste. The present study recommends further research to understand the feasibility of mining waste for the removal of U contamination from groundwater.

Keywords Heavy metals; Geochemical process; Uranium mobilisation; Khetri Copper Belt

Introduction

Uranium (U) is a carcinogenic radionuclide and excessive consumption poses a potential health risk to humans (Blantz et al. 1985). U accumulates in organs and tissues of the human body and causes various health hazards (Russell and Kathren 2004; Wei et al. 2019). It leads to chronic disorders of the liver, kidney and bones (Craft et al. 2004; Brugge and Buchner 2011; Katz 2014). The most important route of U entry is through drinking water. World Health Organisation (WHO) recommends 30 $\mu$g L\textsuperscript{-1} as a permissible limit for drinking water (WHO 2011) exceeding this permissible limit could cause chronic health impacts on human beings.

Sources of ions in groundwater are being controlled by both anthropogenic and geogenic processes. Anthropogenically, contamination of groundwater due to U mines is reported in different countries such as Poland (Kozak et al. 2013), France (Cuvier et al. 2015), Portugal (Neiva et al. 2014), Spain (Villa et al. 2011) and Serbia (Momčilović et al. 2013). A high concentration of U in groundwater is attributed to the abundance of U-rich
mining waste. Simultaneously, some studies suggested geogenic origin of U in groundwater worldwide namely Korea (Shin et al. 2016), USA (Burow et al. 2017), Switzerland (Stalder et al. 2012), Germany (Liesch et al. 2015) and China (Wu et al. 2014).

In India high concentration of U is found in the states of Punjab (Sharma et al., 2019), Himachal Pradesh (Rani et al. 2013), Jharkhand (Patra et al. 2013), Tamil Nadu (Selvi et al. 2016) and Jammu and Kashmir (Sharma et al. 2017). A high concentration of U is observed in different parts of Rajasthan specially in Sikar (Duggal et al. 2016), Jaipur, Ajmer, Tonk, Kota, Jodhpur, and Barmer (Coyte et al. 2019). Hence it is important to understand the geochemical processes governing U mobility in natural conditions.

The high content of uranium is found in the forms of carbonates (Andersonite, Bayleyite and Grimselite), phosphates (Autunite and Torbernite), oxides (Metaschoepite, Metastudtite and Uraninite) and silicates (Uraninite and Swamboite) (Deditius et al. 2008; Gorman-Lewis et al. 2007; Gorman-Lewis et al. 2008a & b; O’Brien and Williams 1983; Ray et al. 2011; Locock and Burns 2003; Gorman-Lewis et al. 2009). Groundwater depletion leads to the dissolution of minerals and enhances elemental mobility. Uraninite is insoluble and is readily oxidized through rock-water interactions to soluble uranyl ions (U(VI)O_2^{2+}) through dissolution, complex formation and desorption-sorption (Tricca et al. 2000). Under anoxic conditions reduced sulfur and iron species may reduce U(VI) to U_3O_8/U_4O_9/UO_2 (Hua and Deng, 2008) (Eq. 1-4).

\[ UO_2^{2+} + FeS \leftrightarrow S^{2-} - UO_2^{2+} + Fe^{2+} \]  --(1)

\[ S^{2-} - UO_2^{2+} \leftrightarrow S^0(s) - UO_2(s) \]  --(2)

\[ FeS(s) + H_2O \leftrightarrow Fe^{2+} + HS^{-} + OH^- \]  --(3)

\[ UO_2^{2+} + HS^{-} \leftrightarrow UO_2(s) - S^0(s) + H^+ \]  --(4)

Natural factors such as geology and geochemical processes such as ion exchange, precipitation, complexation or redox reaction play an important role in releasing the U from minerals to groundwater. Migration of U through sulphide, silicate and carbonate-rich geological setup influence the chemical composition of groundwater. In the mining region dissolution of sulphides contaminates groundwater with a high concentration of heavy metals (Tomiyama et al. 2019; Cánovas et al. 2016). Sulphides specifically Fe, affects the mobility of U by adsorbing on its surface through substitution (Cumberländ et al. 2016) or ion exchange process (Jang et al. 2008). In contrast, Paradis et al. (2016) suggested reduced sulfur-bearing species are not fully effective to limit the mobility of U. In the carbonate-rich region, adsorption of U on soil decreases significantly because of the formation of negatively-charged uranyl carbonate (Giblin et al. 1981).

Geological features such as fractures and faults also create oxidising conditions. Under oxic conditions dissolution of Fe-rich minerals increases and thus in groundwater (Smith and Roychoudhury 2013). Groundwater present in the fracture zone is prone to more contamination. Fracture zones yield most of the water and are also helpful in locating the available water resources (Banks et al., 1992). The iron-bearing minerals adsorb and immobilize the U(VI)-bearing species at circumneutral pH under oxidizing conditions (Li and Kaplan 2012). U gets mobilise in the presence of oxygen and nitrate in groundwater (Watson et al. 2013).

Due to the complex behaviour of U, geochemical processes governing the U mobility in groundwater are difficult to understand. Considering this the region active for mining for different types of minerals such as granite,
quartzite, and copper is selected. The selected study area would be helpful in better understanding the perspectives of U mobility under the influence of different types of anthropogenic and geogenic factors. The main aim is to understand (1) the impact of mining on the concentration of Fe, Mn and U and (2) understanding the perspectives of U mobility under the influence of different types of anthropogenic and geogenic factors.

**Study Area**

The study area is well known for copper mines and it is located in the Jhunjhunu district of the Indian state Rajasthan (Fig. 1a). It is located at an altitude of approximately 370 m above mean sea level with N 28°04.070’ latitudes and E 75°49.294’ longitudes. The region is famous as a Khetri Copper Belt (KBC) since historical times and it extends from Singhana (north) to Raghunathgarh (South) which is~80 km in length. Rocks of Khetri complex are comprises of metasedimentary and igneous intruded by granitoids (Sarkar and Dasgupta 1980; Gupta et al. 1998).

Geologically, the study area falls in the Delhi supergroup and is divided into northern (North Delhi fold belt) and southern (South Delhi fold belt) parts by Kantli fault (NW-SE transverse) (Gupta et al. 1998). Rocks of the supergroup undergoes many folding and polyphase metamorphism (Kaur et al. 2012). The Delhi Supergroup rocks are divided into two groups based on age i.e. an older Alwar (dominate in psammitite) and a younger Ajabgarh group (dominate in pelites) (Heron 1923; Das Gupta 1968). Most of the region is covered by the Ajabgarh group which is formed of metamorphosed argillites and calcareous followed by the Alwar group which dominantly consists of arenaceous. The Ajabgarh group is composed of schists and phyllites and intruded by basic rocks such as granites and amphibolites. The Alwar group is made up of various types of rocks and the composition varies from pure ortho-quartzites to arkose with bands of magnetite, hematite and amphibole. At the confluence of different lithological units (Ajabgarh and Alwar) major faults and fractures are observed (Roy Chowdhury and Das Gupta 1965). The juncture of Alwar and Ajabgarh is also enriched in iron oxide-copper-gold deposits (Knight et al. 2002). The metasomatic alternation of granites in the region are enriched in U and rare earth elements mineralization (Kaur et al. 2014).

**Methodology**

**Sampling**

The groundwater samples were collected in May 2019 following the sampling protocols. The region is famous for copper (Khetri Nagar) mines but quartzite (Ishakpura) and granites (Papurana) mines are also active in the area. The sampling locations were categorized into 3 zones i.e. copper mine zone (CMZ), quartzite mine zone (QMZ) and granite mine zone (GMZ) depending on the type of mine. A total of 32 groundwater samples i.e. 7, 13 and 12 were collected from QMZ, CMZ and GMZ respectively. Considering each mine as a centre point, the sampling locations were distributed systematically in North, East, South and West directions. From each direction, a total of approximately 3 samples were collected after the interval of 2-3 km. The main constraint of sampling was the non-availability of functional hand-pumps. To avoid contamination due to the pipeline the groundwater pumped during the initial first 5 minutes was discarded.
For the perseveration of water samples pH is maintained below 2 by adding few drops of concentrated Supra-pure HNO₃ (Radojevic and Bashkin 1999). After the addition of HNO₃, the samples were filtered using standard pore size 0.45 μm syringe filters at the sampling site. Before the sampling, the sample bottles were washed with Milli-Q ultrapure and the sample itself to avoid contamination. Parameters such as pH, EC and temperature of the samples were measured in-situ using a portable pH meter (Hanna instrument, H196107).

**Elemental and statistical Analysis**

The groundwater analysis for Fe, Mn and radionuclide (²³⁸U) was done using Inductively coupled plasma mass spectrometry (ICP-MS) at Inter-University Accelerator Centre (IUAC), New Delhi. The ICP-MS was calibrated using standard solutions of 1, 20, 50 and 100 μg/L prepared from 1000 μgL⁻¹ multi-element stock solution (1000 μgL⁻¹) via dilution process. For QA/QC, standards namely DGH, MBH, JB-1a and AMH were used (Rathi et Al. 1994; Imai et Al. 1995; Saini et Al. 2013). To assess the precision and accuracy during the analysis elemental concentration in the blank (Milli Q prepared from 18.2 MΩ·cm Ultrapure, Sigma Aldrich) and standards were measured after every 10 samples. The elemental detection limit varies from 0.1-1000 μg/L for Fe, Mn and radionuclide.

The statistical parameters such as average, standard deviation, and median were calculated for the interpretation of data. ArcGIS 10.6.1 was used for locating the sampling locations on the geological map using GPS locations. The inverse distance weighting (IDW) method was used for the multivariate interpolation of U concentration. Graphs were plotted using the Grapher software (version 10).

**Risk assessment**

Drinking water containing excessive U for a longer duration adversely affects kidneys and bones (Zamora et al. 1998; Kurttio et al. 2005). The risk caused by excessive intake of U is calculated in terms of Average Daily Dose (ADD) via equation 5 (WHO 2011):

\[
ADD = \frac{U \times DWI \times EF \times ED}{LE \times BW}
\]

The full form of used abbreviation are as follows U= uranium concentration (mg/L), DWI= Daily Intake of Water (4.05 L per day) (Saini et al. 2016), EF = Exposure Frequency (350 days per year) (USEPA 1999), ED = Exposure Duration (65 years) (WHO, 2011), LE = Life Expectancy (65 years with 365 days per year i.e. 23,725 days) and BW = Body Weight (53 kg of an adult Indian standard person) (Sharma et al. 2017).

Hazard Quotient (HQ) indicates the probable health risk caused by the ingestion of uranium on the consumption of contaminated water. HQ values less than 1 suggest no adverse health effects due to the consumption of uranium contaminated water (AERB 2004) and it was calculated using equation 6.

\[
HQ = \frac{ADD}{RD}
\]

where RD indicates the Reference Dose.

**Results and discussions**
**Variation in concentration of heavy metals and U**

U concentration varies from 0.03 to 430 µg/L with an average of 37 µg/L indicating high contamination in the region. U concentration exceeds or near the WHO permissible limit (30 µg/L) at most of locations. It is observed high at all the sites falling in quartzites mine zone with an average concentration of 87 µg/L. Fe concentration is also observed very high with an average concentration of 2.48±3.31 ppm. Fe exceeds BIS permissible limit (0.3 ppm) at all the sites except two locations.

High concentration of Mn (133 µg/L) and Fe (6.56 mg/L) in the copper mine zone compared to quartzite and granite mine zone is attributed to dissolution of sulphides. The average U concentration is observed lowest (13 µg/L) in the copper mine zone. Baidya and Pal (2020) reported mineralization of uraninite near the Koliyan and Chaandmari Cu deposits confirming the abundance of U in the copper mine zone. Uraninite is the probable source for U in the groundwater. The low concentration of U in groundwater of copper mine zone is attributed to the scavenging capacity of sulphides such as Mackinawite (FeS₉) or Fe-oxyhydroxides (Leermakers et al. 2016; Abdelouas et al. 1999). It also suggests immobilisation of U in groundwater by sulphide dissolution. Thus, the present study confirms that sulphide mines have a buffering capacity to control the U mobilization in groundwater.

**Source of U**

The presence of U-rich granites and albitised quartzite rocks (GSI 2011) are a major source of U in groundwater of the North-Eastern part of Rajasthan. Statistical variation of U concentration is classified into three classes (Fig. 2). The highest value of the first class is 30 µg/L which is a WHO permissible limit and rest are further classified into two classes i.e. high (31-120 µg/L) and severe (above 120 µg/L). In the copper mine zone, U (90 µg/L) concentration is observed high at CE2 located near to the confluence of two geological groups (Ajabgarh and Alwar) compared to other locations. Fractures at the confluence of geological groups create oxic conditions which lead to the dissolution of minerals and permits U mobilization into groundwater (Abdelouas et al. 1999). Adithya et al. (2019) observed that groundwater in the oxidising conditions contain a higher concentration of dissolved U. Faults occur at the juncture of Ajabgarh and Alwar groups are attributed to the high mobility of U in groundwater. The current study is preliminary in nature and further detailed study is needed with more number of samples from the confluence of geological groups. High contamination at ME3 (80 µg/L U) is due to its closeness to albite line. The albite line is enriched in uraninite (Ray, 1990), a potential source of U in the groundwater. ME3 is located in close proximity of Dabla granitoid and illegal mining is also observed in its surroundings. The high concentration of U in the groundwater is also attributed to anthropogenic activities (small scale mines).

MC and MS1 with a U concentration of 216 and 31 µg/L are located close to active granite mines. Sedimentary rocks derived from granites are enriched in uranium concentration (Hobday and Galloway 1999). Almost all the locations in the quartzite mine zone are contaminated with U indicating its high mobility. Quartzite and granite mine zone are located in the vicinity of albite line, where occurrence of U mineralization is reported. Highest concentration of U i.e. 430 µg/L is observed at QW3 close to Gothra granitoid. The high concentration of U is attributed to the presence of uranium enriched granite in the region (GSI 2011; Duggal et al. 2020). The variation in U concentration with respect to their geological settings indicates its geogenic origin in the groundwater.

Classifying the samples according to geological groups, the concentration of Fe and Mn is observed high at the confluence of the Ajabgarh and Alwar groups (Fig. 3). Natural recharge leads to the dissolution of minerals in the
fracture zones. The high concentration of elemental concentration is observed high in the fracture zone due to the presence of the oxic condition. Thus, fractures and faults at the confluence of two geological groups create oxic conditions which enhance the dissolution of Fe-rich minerals. The dissolved Fe and Mn minerals act as a scavenger for U and thus, reducing the U concentration which is supposed to be high at the confluence. At the Alwar group, Fe and Mn concentration are lowest among all the geological setups and U concentration is the highest. It indicates that in natural conditions sulphide mines acts as a buffer and control the dissolution of U in groundwater. In the Khetri region, despite uranium-rich minerals, U concentration is observed low compared to its neighbouring locations, which is due to the buffering capacity of sulphide mines.

In the KBC region, the abundance of uraninite is observed in close vicinity to quartzite and copper mines (Ray 1990; Baidya and Pal 2020). However, a high concentration of U is observed in the groundwater of the quartzite mine zone comparative to the copper mine zone. In the copper mine zone, a high concentration of Fe and Mn is observed and it is well known that the Fe is a scavenger for U in natural conditions. A strong correlation between Fe and U suggests that Fe scavenges U from the groundwater in the copper mine zone.

**Processes affecting U concentration in groundwater**

High U content in groundwater is due to weathering or anthropogenic dissolution of U bearing minerals. The origin of U in groundwater is geogenic but it is also significantly influenced by anthropogenic activities. Both geogenic and anthropogenic factors will be discussed in detail in the following sections.

**Weathering**

A shallow groundwater table with fractures and faults could be the reason for the leaching of U in groundwater. The primary source in the region is U bearing silicate minerals such as granite and uraninite. The region is known for Cu mineralization along with the occurrences of Au ± Fe ± Co ± Ag ± REE ± U ± P (Baidya et al. 2017). The geology of the region is very diverse and made up of different types of parent rocks. Fractures and faults are common features found at the boundary of two-parent rocks. Thus, a high concentration of U in groundwater is attributed to mineral dissolution, faults or fractures in the region.

Groundwater depth is shallow and prone to leaching of elements because of precipitation. U concentration is observed high in shallow groundwater of semi-arid (Coyte et al. 2018) and arid region (Burow et al. 2017) due to leaching. The Khetri belt lies in the semi-arid region with shallow groundwater. The groundwater table is shallow hence weathering of U bearing minerals is a significant contributor of U in groundwater of the Khetri belt. In the Indian scenario, a high concentration of U in groundwater is attributed to an oxidising condition which leads to the solubility of U by forming the soluble uranyl carbonates (Coyte et al. 2017; Brindha and Elango 2013).

**Anthropogenic activities**

Anthropogenic activities (industries or mining) leads to overexploitation or pollution of resources. Granite, quartzite and copper mines are major anthropogenic activities in the region. The concentration of U is found high near quartzite and granite mines due to the abundance of granite minerals. U does not show a significant correlation with Fe (except at the confluence of Ajabgarh and Alwar group) suggesting it is not associate with sulphide rich minerals and its origin is geogenic in the groundwater. At the confluence of Ajabgarh and Alwar group, fractures and faults promote the dissolution of sulphides and U enriched silicates. U and Fe have a
significant correlation coefficient ($R^2=0.97$) at the confluence of Ajabgarh and Alwar group which indicates a strong association between U and Fe. Additionally, fractures and faults create oxic conditions which are responsible for the desorption of U from clay minerals, metal oxides and oxyhydroxides (Zhou and Gu 2005; Yang et al. 2014).

Neighbouring quartzite mines groundwater resources are overexploited and groundwater resources are completely depleted at some locations. Overexploitation is a situation where the rate of groundwater extraction exceeds the average rate of aquifer recharge (Foster and Loucks 2006). A high concentration of U is attributed to overexploitation of groundwater resources. The overexploitation of groundwater leads to an increase in the concentration of major ions (Bagheri et al. 2019) and Fe (Gad et al. 2016). Quartzite mines lie in the Alwar group comprised of quartzite marble, schist and gneiss is a probable source in addition to Gothra granitoid.

**Future research scope**

In the present study, U concentration is observed less in copper mining zone compared to other locations despite the abundance of uraninite or granite, a source of U. It indicates that Fe-hydroxides or FeS are potential scavenger of U from the groundwater. Mine waste enriched in iron sulphides is generated in huge amount across the globe (Thom et al. 2013; Eurostat, 2017). Thus, the present study purposes application of mining waste (tailings and overburden rocks) for the treatment of U concentration from groundwater. The natural attenuation of U from groundwater using mining waste needs further research, as mining waste is also a source of heavy metal contamination.

Mining waste is enriched in heavy metals (Punia et al. 2017), a source of groundwater contamination (Olías et al. 2021). Mining waste highly enriched in FeS generates acid mine drainage (AMD) and contaminates downstream groundwater. Identification of optimum concentration of mining waste for the removal of U from groundwater is important. Achieving a balance between contamination and natural attenuation by mining waste is a topic of debate and needs further research. The dose of mining waste in groundwater after a regular interval of time would be beneficial as excessive FeS would generate AMD. A research is needed to identify the appropriate dose of mining waste and exposure duration to achieve the better removal of U from the groundwater.

The Fe-(hydr)oxides and goethite have a capacity to remove U from groundwater via complexation process (Logue et al. 2004; Gustafsson et al. 2009). Beneficiation of sulphide ore generates Fe enriched mining tailings in abundant quantity and oxidation transforms sulphide minerals into stable goethite (Kim and Kim 2021). Iron oxyhydroxides and goethite controls U mobility in the environment (Lahrouch et al. 2021). Now a day’s permeable reactor barrier (PRB) is widely used technology for the treatment of groundwater. Use of mine tailings in PRB could be a probable solution for the treatment of U contamination. Further research is needed to facilitate the use mine waste as a reactive material for the manufacturing of PRB. The PRB coated with reactive material of Zero-valent iron nanoparticle are found feasible for removal and sequestration of U from groundwater (Li et al. 2015). The injection of iron nanoparticles effectively removes U contamination but the influence of other ions such as calcium, sulphate and carbonate on the water resources needs more studies (Jing et al. 2019).

The huge quantity of mining waste is burden for environment and its use for the treatment of groundwater would solve the problem of its disposal. The preparation of nano particles of Fe also increases the implementation cost at larger scale and the application of mining waste for the same would reduce the cost. Iron sulphide immobilizes
U by reduction or ion exchange processes (Gong et al. 2016; Hua and Deng 2008). Further research is needed to identify the weight% concentration of mining waste which could be used for the removal of U contamination from groundwater. As per our knowledge, no study has been carried for the removal of U contamination from groundwater using mining waste.

**Risk assessment**

Results show that at most of the locations HQ value lies near 1 and at four locations (QW3, CE2, MC, and ME3) it is above 1 indicating the probable adverse health impacts (Fig. 4). The reference dose value is taken as 4.53 mg kg⁻¹ day⁻¹ (AERB 2004). HQ varies from 0 to 7 with an average of 0.61. Consumption of U contaminated water leads to nephrotoxicity, genotoxicity and developmental defects in humans (Brugge and Buchner 2011; Lariviere et al. 2007). Maximum HQ values ~7 demands further studies to assess the health impact of U in the region.

**Conclusions**

The region is active in mining so the impact of anthropogenic activities can’t be neglected. It is observed that sulphide mine acts as a buffer and prevents the dissolution of U-rich minerals in groundwater. U concentration is found high in quartzite mine zone due to lack of buffers i.e. sulphides or Fe-hydroxides. The study recommends immobilization of U in groundwater using mining waste enriched in Fe sulphides and hydroxides. U concentration is observed high near to the confluence of two geological groups compared to other locations. Faults or fractures between two rocks cause oxic conditions and oxygen leads to the dissolution of Fe-rich minerals. Source of U in groundwater is uranium mineralization in granites, gneisses and quartzite rocks. The presence of fractures and faults significantly affects the uranium concentration in groundwater. Shallow aquifers with fractures and faults could be the reason for the mobility of U in groundwater. The origin of U in the groundwater is geogenic in nature but the influence of overexploitation of water can’t be neglected. Overexploitation of groundwater increases the ionic concentration in groundwater. The health risk assessment for cancer indicates the HQ values ~7 demands further studies to assess the health impact of U in the region.

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Figure captions:

Figure 1  Study area C) Sampling locations superimposed on the geology (after Knight et al. 2002; Kaur et al. 2005).

Figure 2  Measured Uranium concentration in the Khetri Copper Belt.

Figure 3  Estimated concentration of heavy metals and U in different geological setups of Khetri region.

Figure 4  Locations with high Average Daily Dose (ADD) and Hazard Quotient (HQ) values.