

# Hydrogeochemical Processes Governing Uranium Mobility: Inferences from the Anthropogenically Disturbed, Semi-arid Region of India

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## Research Article

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# 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  $\mu\text{gL}^{-1}$ ) is observed in the quartzite mine zone whereas minimum (average 13  $\mu\text{gL}^{-1}$ ) 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

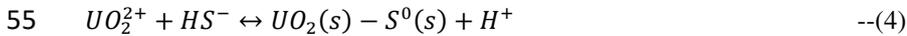
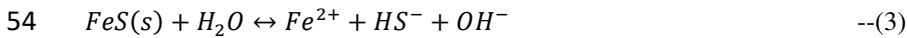
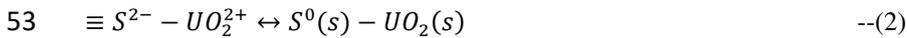
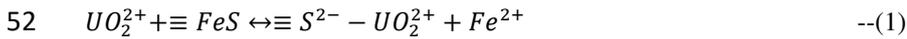
Uranium (U) is a carcinogenic radionuclide and excessive consumption poses a potential health risk to humans (Blantz et al. 1985). U accumulates in organs and tissues of the human body and causes various health hazards (Russell and Kathren 2004; Wei et al. 2019). It leads to chronic disorders of the 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  $\mu\text{g L}^{-1}$  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

36 mining waste. Simultaneously, some studies suggested geogenic origin of U in groundwater worldwide namely  
 37 Korea (Shin et al. 2016), USA (Burow et al. 2017), Switzerland (Stalder et al. 2012), Germany (Liesch et al. 2015)  
 38 and China (Wu et al. 2014).

39 In India high concentration of U is found in the states of Punjab (Sharma et al., 2019), Himachal Pradesh (Rani et  
 40 al. 2013), Jharkhand (Patra et al. 2013), Tamil Nadu (Selvi et al. 2016) and Jammu and Kashmir (Sharma et al.  
 41 2017). A high concentration of U is observed in different parts of Rajasthan specially in Sikar (Duggal et al. 2016),  
 42 Jaipur, Ajmer, Tonk, Kota, Jodhpur, and Barmer (Coyte et al. 2019). Hence it is important to understand the  
 43 geochemical processes governing U mobility in natural conditions.

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



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

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

71 Due to the complex behaviour of U, geochemical processes governing the U mobility in groundwater are difficult  
 72 to understand. Considering this the region active for mining for different types of minerals such as granite,

73 quartzite, and copper is selected. The selected study area would be helpful in better understanding the perspectives  
74 of U mobility under the influence of different types of anthropogenic and geogenic factors. The main aim is to  
75 understand (1) the impact of mining on the concentration of Fe, Mn and U and (2) understanding the perspectives  
76 of U mobility under the influence of different types of anthropogenic and geogenic factors.

77

## 78 **Study Area**

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

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

## 98 **Methodology**

### 99 ***Sampling***

100 The groundwater samples were collected in May 2019 following the sampling protocols. The region is famous  
101 for copper (Khetri Nagar) mines but quartzite (Ishakpura) and granites (Papurana) mines are also active in the  
102 area. The sampling locations were categorized into 3 zones i.e. copper mine zone (CMZ), quartzite mine zone  
103 (QMZ) and granite mine zone (GMZ) depending on the type of mine. A total of 32 groundwater samples i.e. 7,  
104 13 and 12 were collected from QMZ, CMZ and GMZ respectively. Considering each mine as a centre point, the  
105 sampling locations were distributed systematically in North, East, South and West directions. From each direction,  
106 a total of approximately 3 samples were collected after the interval of 2-3 km. The main constraint of sampling  
107 was the non-availability of functional hand-pumps. To avoid contamination due to the pipeline the groundwater  
108 pumped during the initial first 5 minutes was discarded.

109 For the perseveration of water samples pH is maintained below 2 by adding few drops of concentrated Supra-pure  
110 HNO<sub>3</sub> (Radojevic and Bashkin 1999). After the addition of HNO<sub>3</sub>, the samples were filtered using standard pore  
111 size 0.45 µm syringe filters at the sampling site. Before the sampling, the sample bottles were washed with Milli-  
112 Q ultrapure and the sample itself to avoid contamination. Parameters such as pH, EC and temperature of the  
113 samples were measured *in-situ* using a portable pH meter (Hanna instrument, H196107).

### 114 ***Elemental and statistical Analysis***

115 The groundwater analysis for Fe, Mn and radionuclide (<sup>238</sup>U) was done using Inductively coupled plasma mass  
116 spectrometry (ICP-MS) at Inter-University Accelerator Centre (IUAC), New Delhi. The ICP-MS was calibrated  
117 using standard solutions of 1, 20, 50 and 100 µg/L prepared from 1000 µg/L<sup>-1</sup> multi-element stock solution (1000  
118 µg/L<sup>-1</sup>) via dilution process. For QA/QC, standards namely DGH, MBH, JB-1a and AMH were used (Rathi et Al.  
119 1994; Imai et Al. 1995; Saini et Al. 2013). To assess the precision and accuracy during the analysis elemental  
120 concentration in the blank (Milli Q prepared from 18.2 MΩ·cm Ultrapure, Sigma Aldrich) and standards were  
121 measured after every 10 samples. The elemental detection limit varies from 0.1-1000 µg/L for Fe, Mn and  
122 radionuclide.

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

### 127 ***Risk assessment***

128 Drinking water containing excessive U for a longer duration adversely affects kidneys and bones (Zamora et al.  
129 1998; Kurttio et al. 2005). The risk caused by excessive intake of U is calculated in terms of Average Daily Dose  
130 (ADD) via equation 5 (WHO 2011):

$$131 \quad ADD = \frac{U \times DWI \times EF \times ED}{LE \times BW} \quad \text{--5}$$

132 The full form of used abbreviation are as follows U= uranium concentration (mg/L), DWI= Daily Intake of Water  
133 (4.05 L per day) (Saini et al. 2016), EF = Exposure Frequency (350 days per year) (USEPA 1999), ED = Exposure  
134 Duration (65 years) (WHO, 2011), LE = Life Expectancy (65 years with 365 days per year i.e. 23,725 days) and  
135 BW = Body Weight (53 kg of an adult Indian standard person) (Sharma et al. 2017).

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

$$139 \quad HQ = \frac{ADD}{RD} \quad \text{--6}$$

140 where RD indicates the Reference Dose.

### 141 **Results and discussions**

## 142 *Variation in concentration of heavy metals and U*

143 U concentration varies from 0.03 to 430  $\mu\text{gL}^{-1}$  with an average of 37  $\mu\text{gL}^{-1}$  indicating high contamination in the  
144 region. U concentration exceeds or near the WHO permissible limit (30  $\mu\text{gL}^{-1}$ ) at most of locations. It is observed  
145 high at all the sites falling in quartzites mine zone with an average concentration of 87  $\mu\text{gL}^{-1}$ . Fe concentration is  
146 also observed very high with an average concentration of  $2.48 \pm 3.31$  ppm. Fe exceeds BIS permissible limit (0.3  
147 ppm) at all the sites except two locations.

148 High concentration of Mn (133  $\mu\text{gL}^{-1}$ ) and Fe (6.56  $\text{mgL}^{-1}$ ) in the copper mine zone compared to quartzite and  
149 granite mine zone is attributed to dissolution of sulphides. The average U concentration is observed lowest (13  
150  $\mu\text{gL}^{-1}$ ) in the copper mine zone. Baidya and Pal (2020) reported mineralization of uraninite near the Kolihan and  
151 Chaandmari Cu deposits confirming the abundance of U in the copper mine zone. Uraninite is the probable source  
152 for U in the groundwater. The low concentration of U in groundwater of copper mine zone is attributed to the  
153 scavenging capacity of sulphides such as Mackinawite ( $\text{FeS}_{0.9}$ ) or Fe-oxyhydroxides (Leermakers et al. 2016;  
154 Abdelouas et al. 1999). It also suggests immobilisation of U in groundwater by sulphide dissolution. Thus, the  
155 present study confirms that sulphide mines have a buffering capacity to control the U mobilization in groundwater.

## 156 *Source of U*

157 The presence of U-rich granites and albitised quartzite rocks (GSI 2011) are a major source of U in groundwater  
158 of the North-Eastern part of Rajasthan. Statistical variation of U concentration is classified into three classes (Fig.  
159 2). The highest value of the first class is 30  $\mu\text{gL}^{-1}$  which is a WHO permissible limit and rest are further classified  
160 into two classes i.e. high (31-120  $\mu\text{gL}^{-1}$ ) and severe (above 120  $\mu\text{gL}^{-1}$ ). In the copper mine zone, U (90  $\mu\text{gL}^{-1}$ )  
161 concentration is observed high at CE2 located near to the confluence of two geological groups (Ajabgarh and  
162 Alwar) compared to other locations. Fractures at the confluence of geological groups create oxic conditions which  
163 lead to the dissolution of minerals and permits U mobilization into groundwater (Abdelouas et al. 1999). Adithya  
164 et al. (2019) observed that groundwater in the oxidising conditions contain a higher concentration of dissolved U.  
165 Faults occur at the juncture of Ajabgarh and Alwar groups are attributed to the high mobility of U in groundwater.  
166 The current study is preliminary in nature and further detailed study is needed with more number of samples from  
167 the confluence of geological groups. High contamination at ME3 (80  $\mu\text{gL}^{-1}$  U) is due to its closeness to albitite  
168 line. The albitite line is enriched in uraninite (Ray, 1990), a potential source of U in the groundwater. ME3 is  
169 located in close proximity of Dabla granitoid and illegal mining is also observed in its surroundings. The high  
170 concentration of U in the groundwater is also attributed to anthropogenic activities (small scale mines).

171 MC and MS1 with a U concentration of 216 and 31  $\mu\text{gL}^{-1}$  are located close to active granite mines. Sedimentary  
172 rocks derived from granites are enriched in uranium concentration (Hobday and Galloway 1999). Almost all the  
173 locations in the quartzite mine zone are contaminated with U indicating its high mobility. Quartzite and granite  
174 mine zone are located in the vicinity of albitite line, where occurrence of U mineralization is reported. Highest  
175 concentration of U i.e. 430  $\mu\text{gL}^{-1}$  is observed at QW3 close to Gothra granitoid. The high concentration of U is  
176 attributed to the presence of uranium enriched granite in the region (GSI 2011; Duggal et al. 2020). The variation  
177 in U concentration with respect to their geological settings indicates its geogenic origin in the groundwater.

178 Classifying the samples according to geological groups, the concentration of Fe and Mn is observed high at the  
179 confluence of the Ajabgarh and Alwar groups (Fig. 3). Natural recharge leads to the dissolution of minerals in the

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

188 In the KBC region, the abundance of uraninite is observed in close vicinity to quartzite and copper mines (Ray  
189 1990; Baidya and Pal 2020). However, a high concentration of U is observed in the groundwater of the quartzite  
190 mine zone comparative to the copper mine zone. In the copper mine zone, a high concentration of Fe and Mn is  
191 observed and it is well known that the Fe is a scavenger for U in natural conditions. A strong correlation between  
192 Fe and U suggests that Fe scavenges U from the groundwater in the copper mine zone.

### 193 ***Processes affecting U concentration in groundwater***

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

#### 197 **Weathering**

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

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

#### 210 **Anthropogenic activities**

211 Anthropogenic activities (industries or mining) leads to overexploitation or pollution of resources. Granite,  
212 quartzite and copper mines are major anthropogenic activities in the region. The concentration of U is found high  
213 near quartzite and granite mines due to the abundance of granite minerals. U does not show a significant  
214 correlation with Fe (except at the confluence of Ajabgarh and Alwar group) suggesting it is not associate with  
215 sulphide rich minerals and its origin is geogenic in the groundwater. At the confluence of Ajabgarh and Alwar  
216 group, fractures and faults promote the dissolution of sulphides and U enriched silicates. U and Fe have a

217 significant correlation coefficient ( $R^2=0.97$ ) at the confluence of Ajabgarh and Alwar group which indicates a  
218 strong association between U and Fe. Additionally, fractures and faults create oxic conditions which are  
219 responsible for the desorption of U from clay minerals, metal oxides and oxyhydroxides (Zhou and Gu 2005;  
220 Yang et al. 2014).

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

## 227 **Future research scope**

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

235 Mining waste is enriched in heavy metals (Punia et al. 2017), a source of groundwater contamination (Olías et al.  
236 2021). Mining waste highly enriched in FeS generates acid mine drainage (AMD) and contaminates downstream  
237 groundwater. Identification of optimum concentration of mining waste for the removal of U from groundwater is  
238 important. Achieving a balance between contamination and natural attenuation by mining waste is a topic of  
239 debate and needs further research. The dose of mining waste in groundwater after a regular interval of time would  
240 be beneficial as excessive FeS would generate AMD. A research is needed to identify the appropriate dose of  
241 mining waste and exposure duration to achieve the better removal of U from the groundwater.

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

252 The huge quantity of mining waste is burden for environment and its use for the treatment of groundwater would  
253 solve the problem of its disposal. The preparation of nano particles of Fe also increases the implementation cost  
254 at larger scale and the application of mining waste for the same would reduce the cost. Iron sulphide immobilizes

255 U by reduction or ion exchange processes (Gong et al. 2016; Hua and Deng 2008). Further research is needed to  
256 identify the weight% concentration of mining waste which could be used for the removal of U contamination from  
257 groundwater. As per our knowledge, no study has been carried for the removal of U contamination from  
258 groundwater using mining waste.

### 259 ***Risk assessment***

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

### 265 **Conclusions**

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

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487 **Figure captions:**

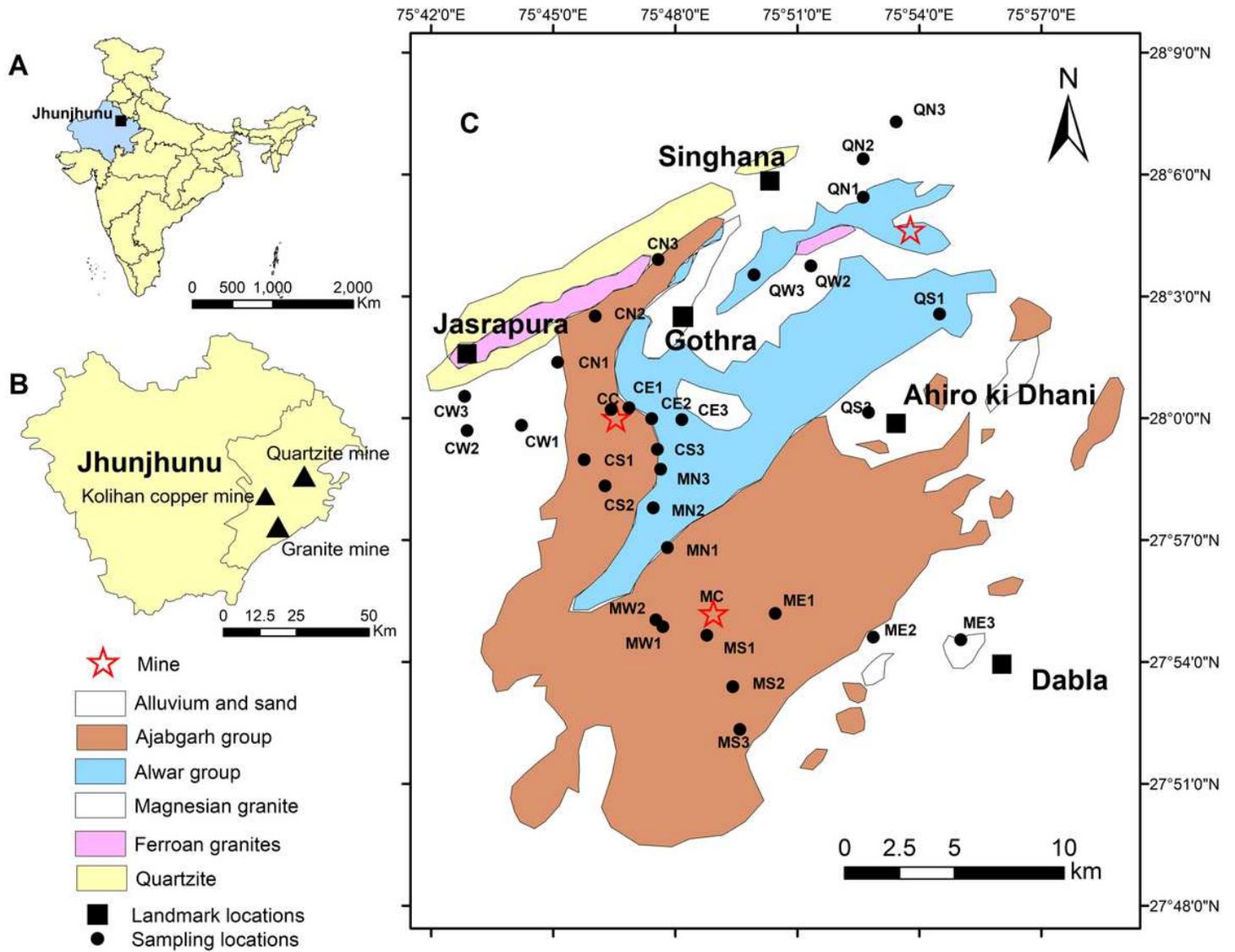
488 Figure 1 Study area C) Sampling locations superimposed on the geology (after Knight et al. 2002; Kaur et al.  
489 2005).

490 Figure 2 Measured Uranium concentration in the Khetri Copper Belt.

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

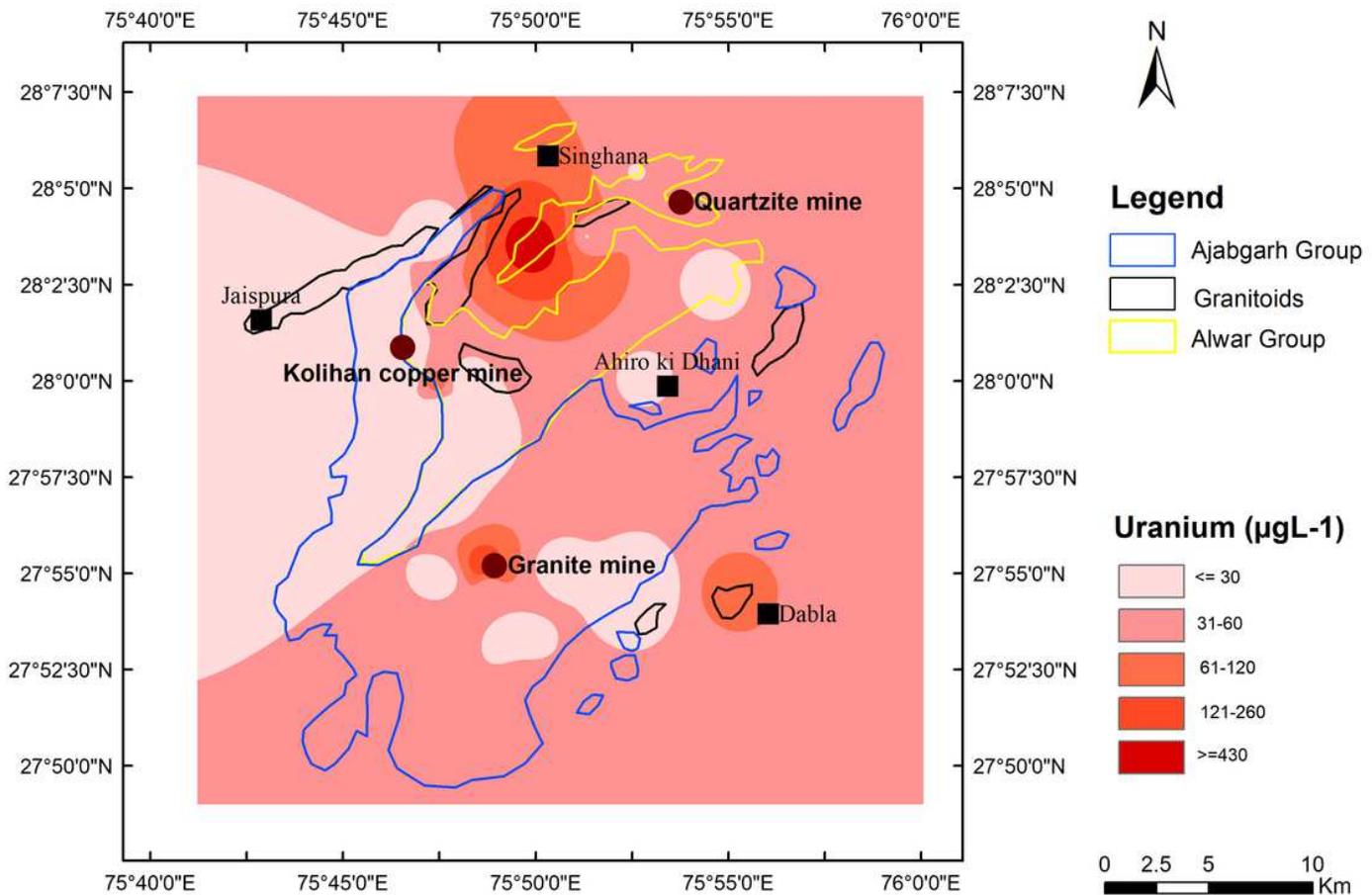
492 Figure 4 Locations with high Average Daily Dose (ADD) and Hazard Quotient (HQ) values.

# Figures



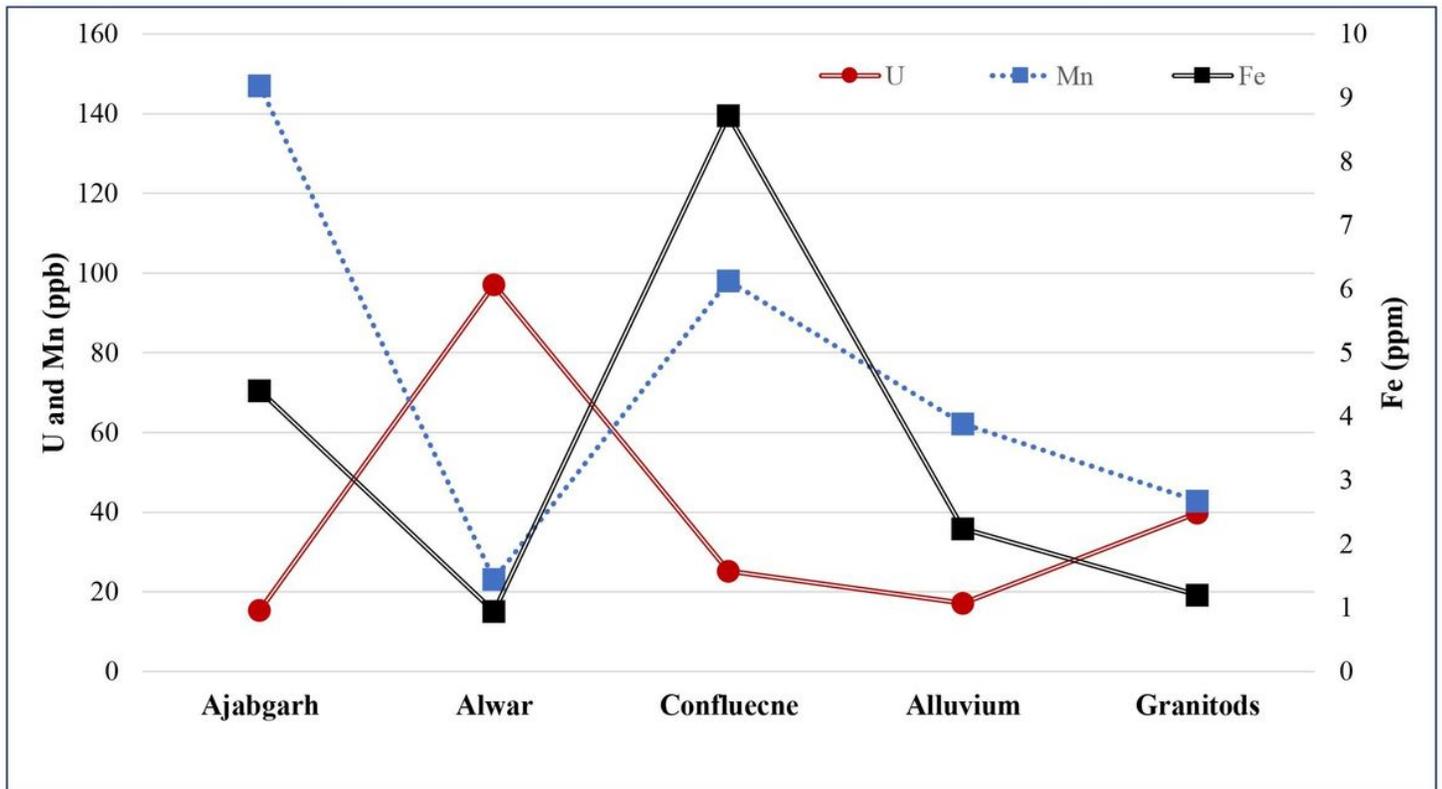
**Figure 1**

Study area C) Sampling locations superimposed on the geology (after Knight et al. 2002; Kaur et al. 2005). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



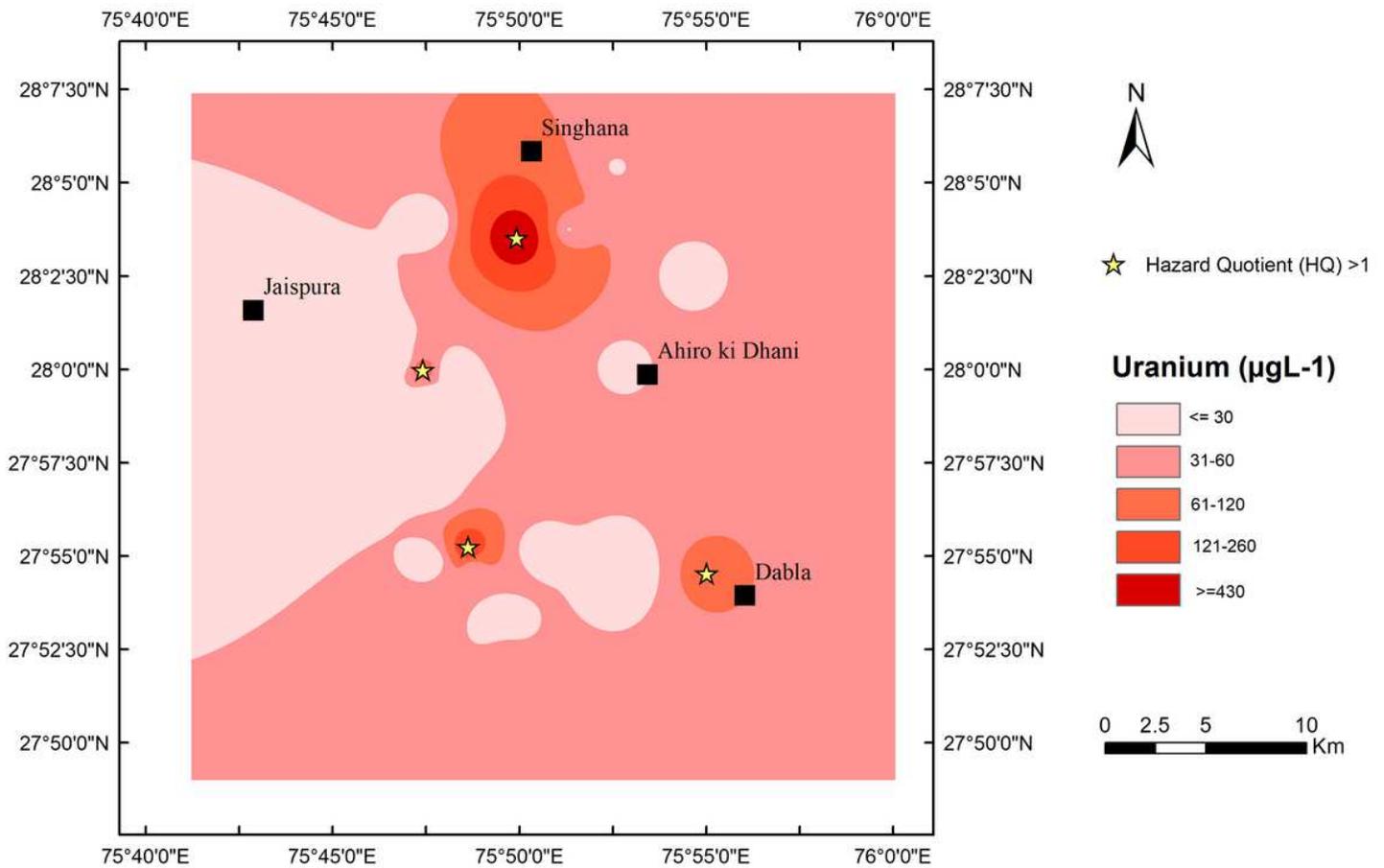
**Figure 2**

Measured Uranium concentration in the Khetri Copper Belt. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



**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. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.