



# Role of colloid in metal transport in river water around Jaduguda uranium mines, Singhbhum shear zone

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This paper attempts to identify and characterize the colloids present in the river water around the Jaduguda uranium mines located in Jharkhand, India in order to understand their role in metal transport. The stream receiving direct release of effluent water from the mine tailing is contaminated and the concentration of Mn, F<sup>-</sup>, Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and U is above the maximum permissible. The colloids mostly consist of Mn-oxide, kaolinite, halloysite and Fe(oxy)hydroxide. The concentration of As, Co, Cu, Mn, Ni, Pb, U and Zn in surface water is very much lower than that of the colloids. This is attributed to the importance of colloids in metal scavenging due to more reactive surface sites. The high resolution transmission electron microscope (HRTEM) study further indicates that Mn-oxides play a most important role in retention and transportation of U in the study area.

**Keywords.** Colloid; uranium; Jaduguda; uranium mines.

## 1. Introduction

Colloids are defined as the particles with a mean diameter of less than 0.45 μm, or a size ranging from 1 nm to 1 μm (Mohamed and Paleologos 2017). Colloids in natural water usually consist of clay, iron (oxyhydro)oxides, organic matter and bacteria (Kretzschmar and Schäfer 2005). With respect to their chemical properties, colloids are often defined as charged species and solids having large specific surface areas (Buffle and Leppard 1995). Therefore, colloids have more affinity to adsorb metal ions at the surface due to their high reactivity. The ability of colloids to facilitate contaminant transport depends on the mobility of these particles and their ability to retain contaminants. The small size range of the colloids implies that they have longer settling times in water and can travel

longer distance in suspension. Thus, they have a tendency to carry contaminants over longer distance (McCarthy and Zachara 1989). Various types of colloidal particles have shown different adsorption capacities for metals. Thus, the fate of reactive elements and/or pollutants is governed by types of colloidal particles in aqueous system (Pédrot *et al.* 2008).

Role of colloids in transport of trace metals along with radionuclides has been reported in many parts of the world (Short *et al.* 1988; Dearlove *et al.* 1991; Zänker *et al.* 2000). Hence, colloids represent potentially important transport vectors for highly insoluble or strongly sorbing metals and radionuclides in the environment. Elements associated with colloids are more bioavailable (John and Leventhal 1995). So, for impact assessment in aquatic ecosystem around the mines, information regarding distribution of metals in dissolved along

with their association with the colloids is very essential.

Several studies have reported the concentration, distribution and effects of radionuclides in the effluents discharged from the Jaduguda mines and tailing (Markose *et al.* 1993; Tripathi *et al.* 2008; Singh and Soni 2010; Sahu *et al.* 2014). According to studies by Raghavayya (1999) and Sethy *et al.* (2013), the concentration of  $^{226}\text{Ra}$  in the Gara river and Subarnarekha river is within limits even in the downstream region of the mines due to high efficiency of treatment in the effluent treatment plant (ETP). Several studies have focused on the occurrence of U and heavy metal around the Jaduguda uranium mines (Tripathi *et al.* 2008; Giri *et al.* 2010; Sethy *et al.* 2013). However, no study has been attempted so far to study the colloids present in the river water and their importance in metal retention around the uranium mines. Thus, this paper proposes to identify the minerals present in colloids and their role in metal transport.

## 2. Study area

Jaduguda uranium mines are located in the Jaduguda district of Jharkhand, India. The mining activities are carried by the Uranium Corporation of India Ltd (UCIL). The mines are situated in a valley of uranium copper mineralized hill of Singhbhum located in the central sector of Singhbhum Shear Zone (Krishna and Rao 1980). Uranium deposit was discovered in 1951 (Gupta and Sarangi 2005). The autoclastic conglomerate and quartz-chlorite-apatite-tourmaline-magnetite schist are the host rocks in which uranium bearing fine grained uraninite occur as disseminated grains and micro-veinlets (Sarangi 2003). Pyrite, chalcopyrite, millerite, pentlandite, molybdenite and magnetite are the major accessory minerals associated with uranium mineralization (Pal *et al.* 2011; Sarkar 1982). The concentration of  $\text{U}_3\text{O}_8$  in the host rock varies from 0.067 to 0.2% (Mahur *et al.* 2008). Uranium mines have an ore processing plant having capacity of about 2100 tonnes per day

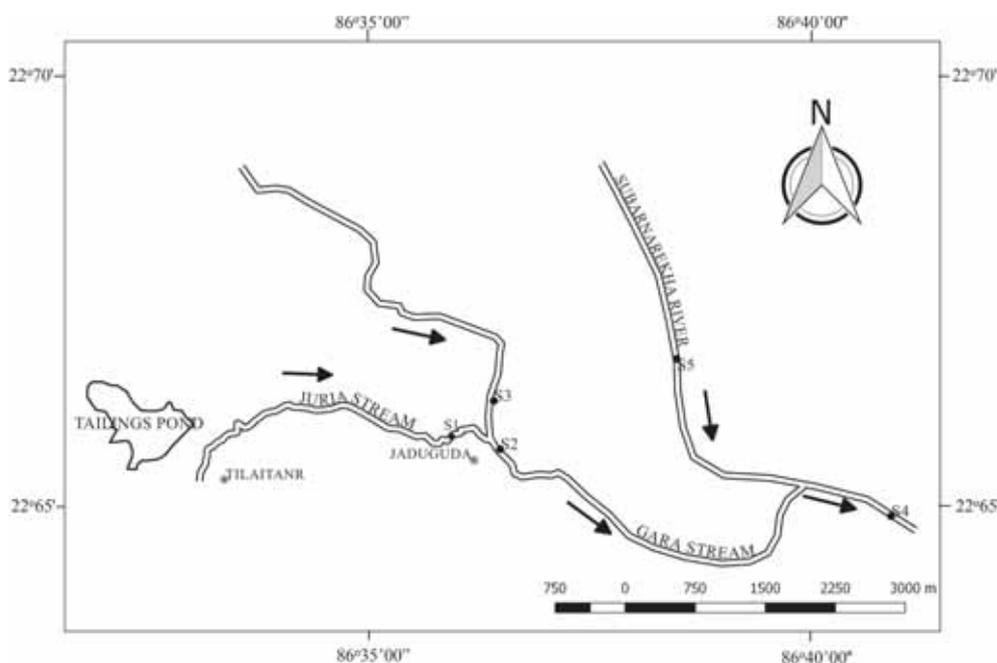


Figure 1. Sample location map.

Table 1. Detection limit (Unit: ppb) and precision of measured cations, anions and metals.

| Element         | $\text{Ca}^{2+}$ | $\text{K}^+$ | $\text{Mg}^{2+}$ | $\text{Na}^+$ | $\text{Cl}^-$ | $\text{F}^-$ | $\text{SO}_4^{2-}$ | Mn   | Zn   | Co   | Ni   | Cu   | As   | Pb   | U    |
|-----------------|------------------|--------------|------------------|---------------|---------------|--------------|--------------------|------|------|------|------|------|------|------|------|
| Detection limit | 6.0              | 7.0          | 7.0              | 5.0           | 20.0          | 5.0          | 20.0               | 0.08 | 0.16 | 0.50 | 2.30 | 0.35 | 2.15 | 1.05 | 0.20 |
| Precision (%)   | 0.5              | 4.0          | 2.8              | 2.6           | 3.5           | 1.1          | 3.2                | 0.98 | 1.05 | 1.45 | 0.95 | 0.80 | 1.10 | 0.85 | 0.91 |

Table 2. Physico-chemical parameters and concentration of various elements in river water around Jaduguda uranium mines.

| Sample | ppm  |      |     |                  |                 |                |                  |                |                 |                               | ppb              |                               |      |      |      |       |      |      |      |       |
|--------|------|------|-----|------------------|-----------------|----------------|------------------|----------------|-----------------|-------------------------------|------------------|-------------------------------|------|------|------|-------|------|------|------|-------|
|        | pH   | EC   | Eh  | Ca <sup>2+</sup> | Na <sup>+</sup> | K <sup>+</sup> | Mg <sup>2+</sup> | F <sup>-</sup> | Cl <sup>-</sup> | SO <sub>4</sub> <sup>2-</sup> | SiO <sub>2</sub> | HCO <sub>3</sub> <sup>-</sup> | Mn   | Zn   | Co   | Ni    | Cu   | As   | Pb   | U     |
| S1     | 7.45 | 2331 | 156 | 3.05             | 217.56          | 23.82          | 0.01             | 3.10           | 185             | 1183                          | 4.19             | 65                            | 2.88 | 0.23 | 1.51 | 17.79 | 4.33 | 1.69 | 3.79 | 32.88 |
| S2     | 8.40 | 292  | 131 | 0.04             | 19.92           | 4.83           | 0.38             | 0.34           | 11.46           | 21.23                         | 7.75             | 110                           | 0.01 | 0.15 | 0.10 | 2.19  | 1.78 | 0.86 | 2.45 | 0.45  |
| S3     | 7.76 | 593  | 154 | 14.5             | 37.33           | 6.03           | 0.63             | 0.63           | 65.54           | 152.41                        | 8.46             | 110                           | 0.20 | 0.24 | 0.28 | 4.52  | 2.33 | 1.22 | 3.95 | 3.4   |
| S4     | 8.01 | 335  | 152 | 0.83             | 20.44           | 4.16           | 1.40             | 0.44           | 20.95           | 24.09                         | 7.53             | 120                           | 0.41 | 0.20 | 0.51 | 3.60  | 3.8  | 0.73 | 4.49 | 0.64  |
| S5     | 8.62 | 249  | 141 | 0.07             | 19.18           | 6.28           | 0.01             | 0.41           | 16.92           | 16.10                         | 5.73             | 80                            | 0.00 | 0.22 | 0.15 | 3.41  | 2.53 | 0.86 | 3.39 | 0.52  |

Unit: EC:  $\mu\text{S}/\text{cm}$ ; Eh: mV; Na-Zn: ppm; Co-U: ppb.

and 80% recovery of uranium (Curtis 2007). The wastes generated after processing of ore are treated in the ETP and released to the nearby the Juria stream through a concrete drain.

### 3. Methodology

The river water and colloid samples were collected from the Juria, Gara and Subernarekha river which are traversing nearby uranium mines. The sample location map is given in figure 1. The water samples were collected in duplicates and filtered through 0.22  $\mu\text{m}$  membrane filter paper *in situ*. One part was acidified with ultra-pure HNO<sub>3</sub> to bring pH < 2 and preserved for analyses cations and metals. The non-acidified portion was used for determination of pH, electrical conductivity (EC) and redox potential (Eh) by ion selective electrodes (ORION model 1260), and anions. The pH, EC and Eh were measured *in situ* by using respective electrodes (Orion). Prior to the analysis of pH, EC and Eh, respective electrodes were calibrated and checked with respective standard solutions. Alkalinity was measured in field by following titration method (APHA 2012). Concentration of anions (Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>), cations (Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>) and silicate was determined using ion chromatography (883 Basic IC Plus, Metrohm). The concentration of metals (Ni, Cu, Co, Mn, Pb, Zn and U) was determined using ICP-MS. Prior to analysis, the ion chromatograph and ICP-MS were calibrated using multi-element standards. The precision and accuracy of the analytical procedures was maintained by analysing blank, duplicate samples and multi-element stock standard solutions. For ICP-MS, standard solutions of 10, 100 and 1000  $\mu\text{g}/\text{L}$  were prepared from stock solution (10 mg/L) using Milli-Q water and ultra-pure grade of nitric acid. The analysis errors were within 5% for all analysis. The analytical precision

Table 3. Concentrations of metals (mg/kg) in colloids collected from the mentioned location.

| Element | S1     | S2     | S4     |
|---------|--------|--------|--------|
| Co      | 105.70 | 54.47  | 29.11  |
| Ni      | 761.43 | 128.31 | 75.06  |
| Cu      | 177.22 | 105.80 | 70.77  |
| Zn      | 251.04 | 154.53 | 117.38 |
| As      | 70.90  | 41.72  | 28.91  |
| Pb      | 10.92  | 27.35  | 30.14  |
| U       | 82.68  | 19.79  | 4.09   |

and detection limit of measured cations, anions and metals are given in table 1.

The colloid samples were collected from S1, S2 and S4 locations. At other two locations, it was not possible to collect the colloids due to their low concentration. Thus, this study is based on the limited number of samples, and may not be representative of the whole area. Water samples were processed in the laboratory within 12 hours of the sampling using tangential-flow ultrafiltration technique (Spectrum, USA), with membranes of nominal cut-offs at 0.2  $\mu\text{m}$  and 5 kD. All membranes were cleaned in the laboratory with Milli-Q water. The fraction between 0.2  $\mu\text{m}$  and 5 kD was

separated and air-dried in the laboratory. This colloid fraction was used for further analysis using high resolution transmission electron microscope (HRTEM).

Morphology, mineral identification and chemical composition of colloids were also carried out using HRTEM (FEI Tecnai G2 F30 S-TWIN) operated at 300 kV. The colloid samples were dispersed in ethanol and drop casted on carbon-coated Cu grid used for HRTEM analysis. The energy dispersive X-ray (EDX) and elemental mapping by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) were done by using Quantax 200 ESPRINT 1.9 version.

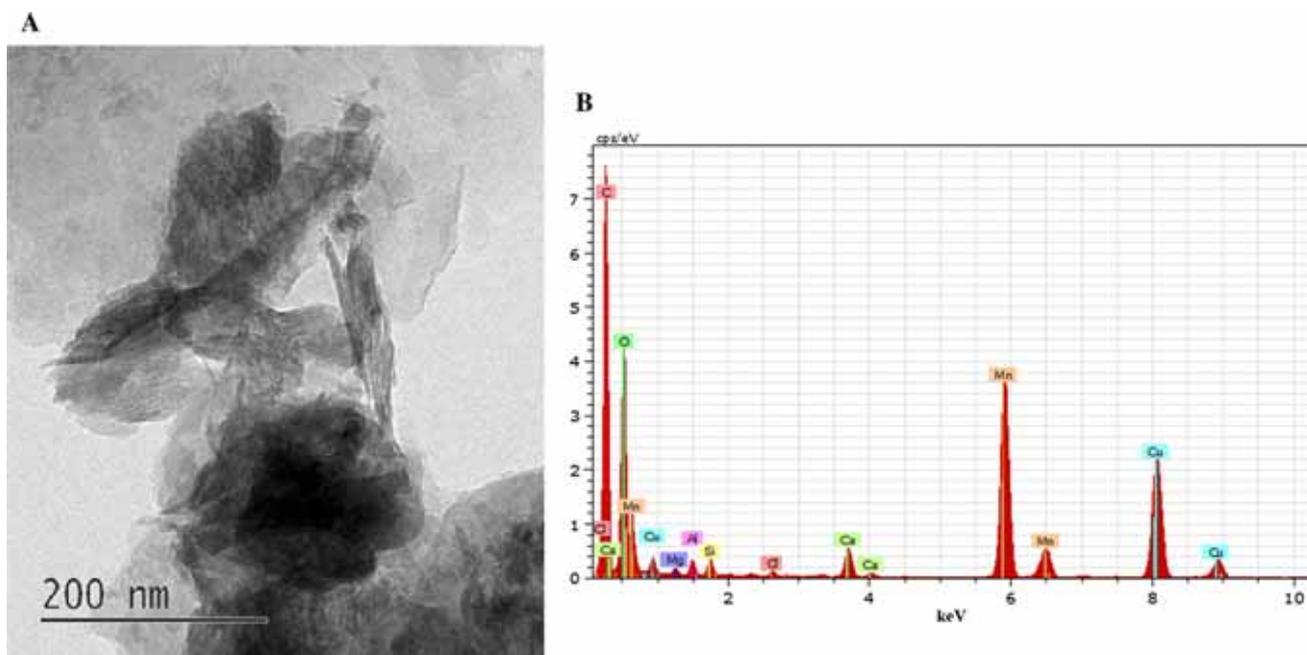


Figure 2. HRTEM image of Mn-oxide (A) and its EDX spectrum (B) collected from S1 location.

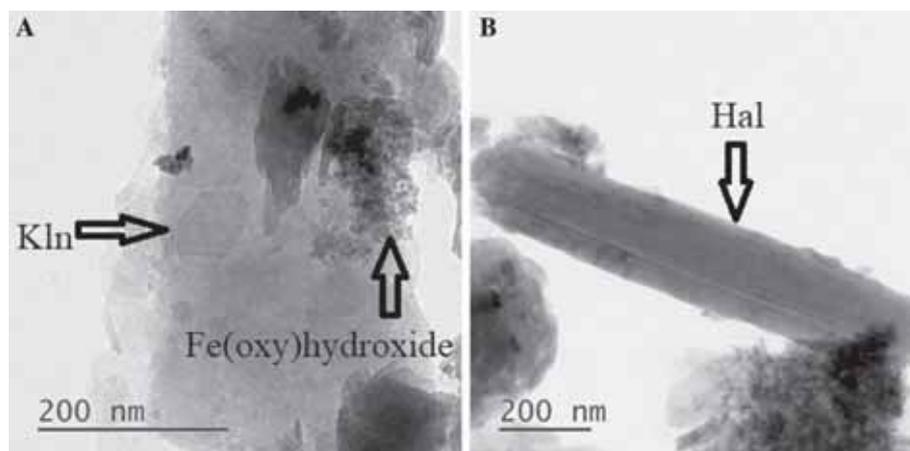


Figure 3. HRTEM image of kaolinite and Fe(oxy)hydroxides (A) and rod shaped halloysite (B) collected from S4 location.

#### 4. Results and discussion

The physico-chemical properties of water samples are given in table 2. The pH of the river water is alkaline in nature as it varies from 7.76 to 8.62. The Eh varies from 130.9 to 155.1 mV. Electrical conductivity ranges from 248.6 to 2331  $\mu\text{S}/\text{cm}$  in the surface water samples. The Juria stream water shows maximum conductivity. It is due to the presence of higher amount of dissolved ions from the effluent discharged by the mine tailing. Maximum concentration of  $\text{SO}_4^{2-}$  (1182 mg/L), Na (218 mg/L) and EC (2331  $\mu\text{S}/\text{cm}$ ) is observed in the Juria stream. It is due to the direct discharge of treated water, which has more dissolved ions, from the tailing pond of the Jaduguda uranium mines. The concentration of  $\text{Na}^+$ , Mn,  $\text{F}^-$ , and  $\text{SO}_4^{2-}$  in the Juria stream exceeds the maximum permissible limits of drinking water (WHO 2017). However, all the elements are within limit in the other river water around the mines. The high concentration of  $\text{SO}_4^{2-}$  and Mn is caused due to their release from the tailing pond after the acid leaching of minerals containing uranium using  $\text{H}_2\text{SO}_4$  in the presence of pyrolusite as oxidant. Also high concentration of  $\text{Na}^+$  is due to utilization of NaCl during uranium beneficiation. The high level of  $\text{F}^-$  in the Juria stream is might be due to its release from the host rock during acid leaching. The apatite, tourmaline and mica present in the host rocks are known to contain high level of F (Bailey 1977), and also release of F is enhanced under acidic condition (Borgnino *et al.* 2013). The concentration of U varies from 0.45 to 32.88  $\mu\text{g}/\text{L}$  though it exceeds the maximum permissible limit (30  $\mu\text{g}/\text{L}$  as prescribed by WHO 2017) in the Juria stream. Tripathi *et al.* (2008) reported that U concentration in the surface waters near the mines is in the range of 0.3–54.9  $\mu\text{g}/\text{L}$ . The decrease in the concentration of elements in surface water of the Gara and Subernarekha river with respect to the that of S1 can be explained by the four mechanisms as suggested by Chapman *et al.* (1983) such as (1) dilution by river water and groundwater seeps downstream, (2) precipitation and subsequent sedimentation of solids, (3) adsorption of dissolved species onto newly-formed solids, (4) adsorption of dissolved species onto existing suspended materials such as clay.

Total concentration of metals and U in the colloids is given in table 3. Uranium concentration associated with colloids varies from 4.09 to 82.68 mg/kg. Similarly, Co ranges from

29.1 to 105.7 mg/kg, Ni 75.1–761.4 mg/kg, Cu 70.8–177.2 mg/kg, Zn 117.4–251.0 mg/kg, Pb 10.9–30.1 mg/kg. Metals associated with colloidal forms have been found in significant amounts compared to that of river water. This shows that colloidal particles can scavenge the metals and U effectively, and travel longer distance due to their lighter weight.

The HRTEM study along with EDX, indicate that colloids mostly consist of Mn-oxide, clay minerals (halloysite and kaolinite), and Fe-oxyhydroxides. The HRTEM image and EDX spectra of Mn-Oxide are shown in figure 2. The Mn-oxides

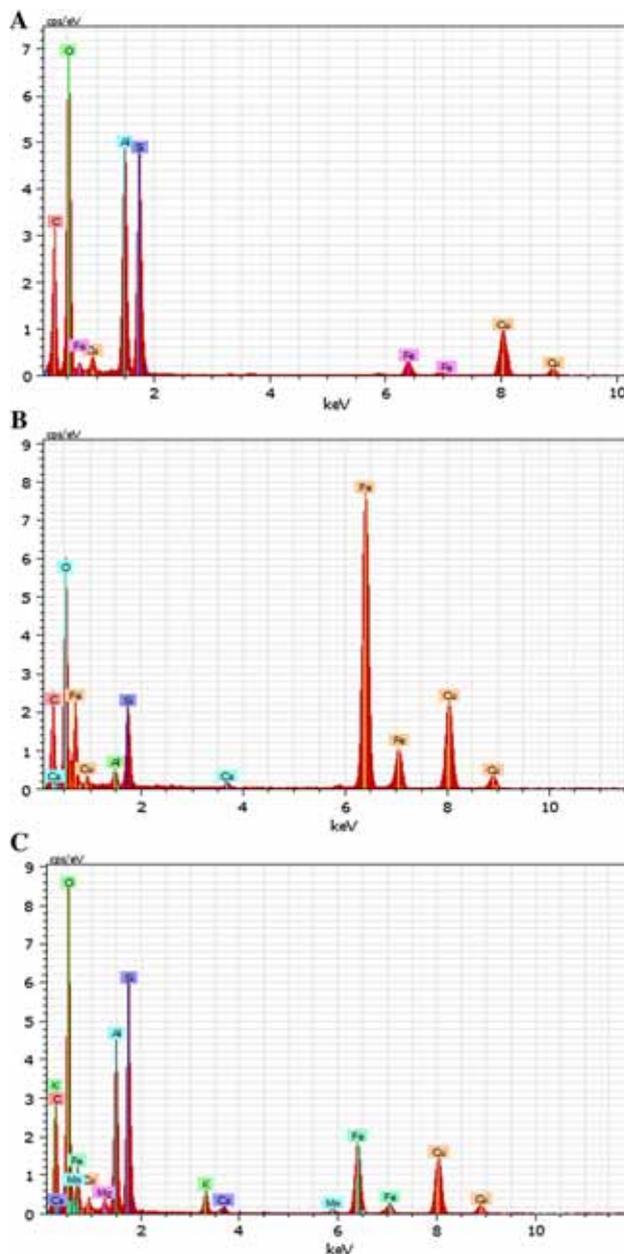


Figure 4. Represents the EDX spectrum of halloysite (A), Fe-oxyhydroxide (B) and kaolinite (C).

show whisker-shaped particles and they mostly agglomerated. Individual whisker varies in width 5–11 nm having length 75–187 nm. The kaolinite and halloysite are identified based on their shape along with EDX spectra. Kaolinite shows pseudo-hexagonal shape (figure 3A), whereas halloysite appears in rod shape (figure 3B). Halloysite is further confirmed from the EDX spectra based on the Al/Si ratio (0.98) which is close to 1, whereas in kaolinite this ratio is 0.73 (figure 4). Reports of occurrences of such colloids in rivers around mines have been reported worldwide (Kimball *et al.* 1995; Schemel *et al.* 2000; Hill and Aplin 2001).

To further, understand the association and distribution of metals and U on the colloids, the HAADF-STEM analysis was conducted. The HAADF-STEM technique is emerged in recent years as a very effective tool to study the composition of crystalline nanoparticles with sub-angstrom

resolution (Johnston-Peck *et al.* 2013; Kauko *et al.* 2013; Woo and Gulians 2016). The HAADF-STEM imaging is a mapping technique which detects the elastically scattered electron by the sample through high angle. Detailed interpretation of the HAADF-STEM tomographic image provides information on the distribution of elements on the surface of nanoparticles that help to understand the metal association. The minerals can also be confirmed based on their chemical mapping in the HAADF-STEM images (figures 5 and 6). It is seen that occurrence of U is observed with Mn-oxides and clay minerals. The association of U with Mn-oxide is much more than other minerals. Occurrence of U with Fe(oxy)hydroxides is not observed through HAADF-STEM image. McKenzie (1980) and Wang *et al.* (2012) reported that Mn-oxides have higher sorption affinity for U than iron oxides with similar surface. Based on experimental studies, Bachmaf

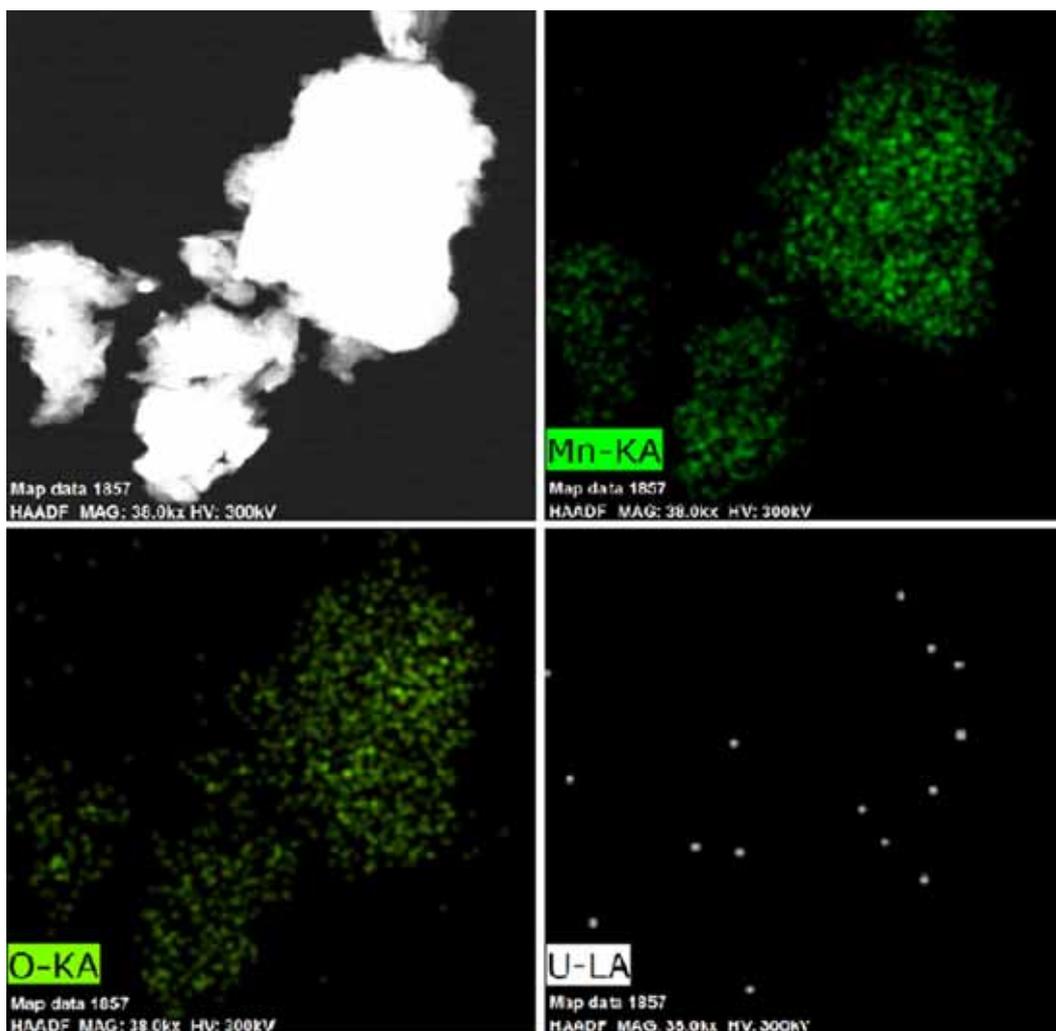


Figure 5. HAADF-STEM image of Mn-oxide and its elemental mapping.

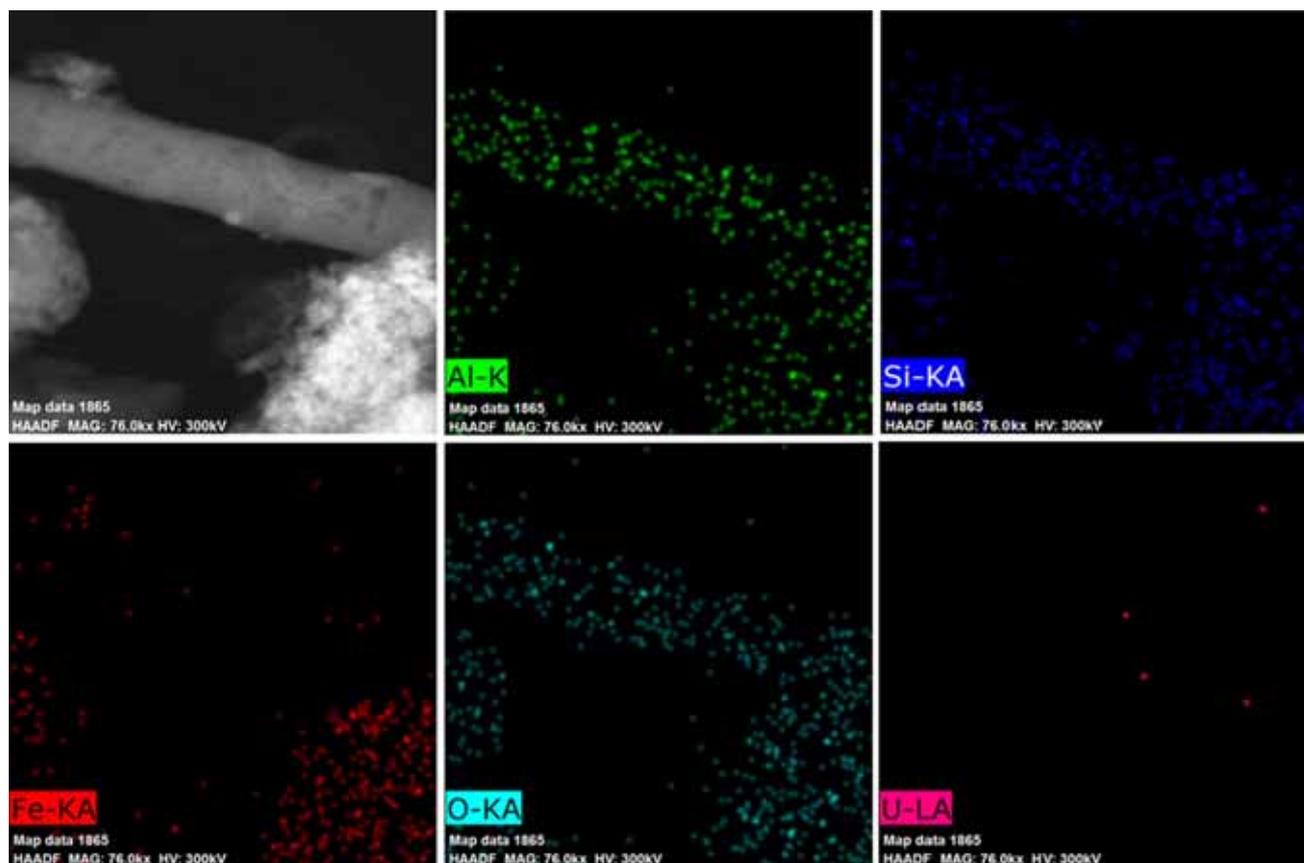


Figure 6. HAADF-STEM image of halloysite and Fe(oxy)hydroxides along with their elemental mapping.

and Merkel (2011) showed that adsorption of U on kaolinite is more than other clay minerals due to more readily available aluminol sites. However, halloysite has been observed to have higher adsorption capacity among other common clay minerals due to its reactive tubular structure (Ferrante *et al.* 2017; Hermawan *et al.* 2018).

## 5. Conclusions

This work presents the concentration of major cation, anion and trace elements in the nearby streams around the uranium mines of, Jaduguda. The colloids present in the surface water are also identified using HRTEM. The stream water is mostly alkaline in nature. The stream receiving effluent from tailing show very high EC due to presence of high dissolved ions; and show high concentration of  $\text{Na}^+$ ,  $\text{F}^-$ , Mn,  $\text{SO}_4^{2-}$  and U. The concentration of U in stream water varies from 0.45 to 32.88  $\mu\text{g}/\text{L}$ . The Mn oxides, clay minerals like halloysite and kaolinite, and Fe-oxyhydroxides are found in the form of colloids. Most of the U is associated with Mn-oxide which plays a dominant

role in mobility of U. The study stresses on the importance of colloids in metal accumulation and transport in the surface water.

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