# Interaction of Uranium Contaminated Groundwater with Alluvial Soils of Punjab

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**Abstract:** The groundwater quality of south-west region of Punjab (i.e. Malwa region) has been marred by the presence of high levels of uranium. Scientific literature suggests that the contamination is attributed to geogenic sources of uranium arising from the Siwalik region. Although many studies have mapped the regions of high levels of uranium contamination studies on the interaction of aqueous uranium with the alluvial soils of Punjab has been lacking. In this study, we focus on understanding the interaction of aqueous uranium in groundwater with the alluvial soils of Punjab region. Both batch kinetic and isotherm studies were performed to decipher the rate of uranium adsorption to soils and the underlying sorption mechanism. Moreover, the variation in adsorption with respect to pH was obtained using pH edges studies. The results of this study will aid in better understanding of geochemical controls of aqueous uranium and its mobility in the groundwater of Punjab.

**Keywords:** Groundwater contamination, Uranium, Malwa region, Adsorption studies

## 1. Introduction

The presence of dissolved uranium in groundwater can be due to geogenic as well as anthropogenic activities.Aqueous uranium is found to be in its hexavalent form in oxic environment.In aqueous solutionuranium (VI) exists as complexesof the stable linear uranyl ion, $UO_2^{2+}$, which forms numeroushydrolysis species (Duff et al.,1996).Sorption interactions with sediments, soils and rocks are very important mechanisms for attenuating the mobility of uranium throughthe subsurface environment (Pabalan et al., 1996).Uranium contamination poses a threat to surface and ground waters (Fan et al., 2011). The groundwater samples of Malwa region of Punjab has been reported to have significantly high levels of uranium. Through literature survey, mapping of uranium contamination in groundwater was done using ArcGIS (Figure 1).

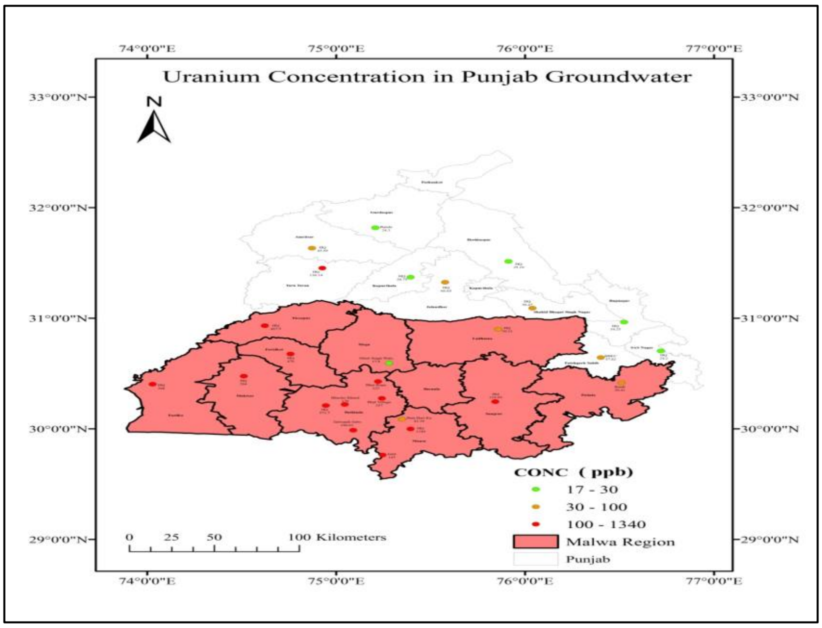

**Fig.1:** Uranium Contamination in Groundwater of Punjab

Migration andretardation of uranyl ions in geological environments arecontrolled mainly by the sorption of these toxic speciesto mineral surfaces (Bachmaf et al., 2008).Hence, predicting the fate and transportof uranyl on contaminated sites requires an understandingof the factors affecting their sorption onto minerals (Bakatula et al., 2015).Previous studies have attributed the occurrence of uranium in groundwater to geogenic processes related to Siwalik sediments (Patnaik et al., 2016). The guideline value for uranium in drinking water as per theWorld Health Organisationis 30 µg/L (WHO). Uranium adsorption onto minerals surfaces play a key role in controlling its mobility in groundwater (Wan et al., 2011).The mobility of U(VI) is of great concern in Malwa region groundwater. The prediction of U(VI) mobility requires understanding the mechanism of U(VI) adsorptionby the soil using a surface complexation model (Davis et al., 2004).

## 2. Objectives

The objective of the study was to investigate the interaction of aqueous uranium with alluvial soils of Punjabunder field relevant scenario by performing adsorption studies. Both U(VI) adsorption kinetics and isotherms experiments were performed. Further, the variation of adsorption as a function of pH was also studied.

## 3. Material and Methodology

The soil samples were collected near the Sutlej River from non-agricultural land which was free of pesticides contamination. The soil was collected at a depth of 90 cm below ground level. After removing debris, grass roots, and pebbles it was left for air drying for 48 hrs. Air dried soil was homogenised, passed through 2-micronsieve and used in the experiments.The pH of soil was determined using ASTM protocol (ASTM method4972-01). Soil pH was measured both in distilled water and 0.01 M calcium chloride solution.

### Reagents and Standards

All chemicals and reagents used for experiment were of ACS certified analytical grade. 10 ppm of uranium working stock solution was prepared from 100 ppm ICP standard solution through dilution.

### Batch Experiments

Batch kinetic and isotherm studies were performed to decipher the rate of uranium adsorption to soils and underlying sorption mechanism. Kinetic studies were performed for determining the duration necessary to achieve equilibrium solution concentration of U(VI) on the basis of which further isotherm studies were performed. All experiments were performed in an electrolyte solution having ionic strength of 0.01 M $NaHCO_3$ and 0.01 M $NaNO_3$that is representative of the groundwater chemistry of uranium contaminated region of Punjab (Hundal, H.S., 2011). The experiments were performed at a soil to solution ratio of 25 g/L. All experiments were performed in duplicates that include control (no soil) and blank samples (no uranium). At specific intervals the test tubes were recovered and centrifuged for 15 minutes at 6000 rpm. After centrifugation,the supernatant was recovered for alkalinity and aqueous uranium determination.Uranium was analysed using the LED Fluorimeter(QuantalasePvt. Ltd.). Alkalinity of samples was measured by titration using 0.02 N $H_2SO_4$.In the case of isotherm studies, variousinitial concentrations of uranium were spiked (0.25 ppm to 1.25 ppm). After an equilibrium time of 48 hours which was found to be sufficient based on kinetic study results, all test tubes were recovered and centrifuged for 15 minutes at 6000 rpm. After centrifugation, supernatant was recovered from each of the test tube for alkalinity and uranium concentration analysis as described above. The difference between the initial and the aqueous concentration of U(VI) was attributed to the adsorption of U(VI)onto soil. Further, the variation in adsorption with respect to pH was obtained using pH edges studies conducted at an initial uranium concentration of 1 ppm by varying the pH between 4.5 to 10.5.

## 4. Results

The ambient pH of the soil samplewas found to be alkaline lying betweenpH 8 and pH 9.The results of the kinetic study(Fig. 2) indicated a sorption equilibrium time of 48 hours. The alkalinity of the system varied between 497 mg/L to 548 mg/L wherein increase in contact time resulted in an increase of alkalinity.

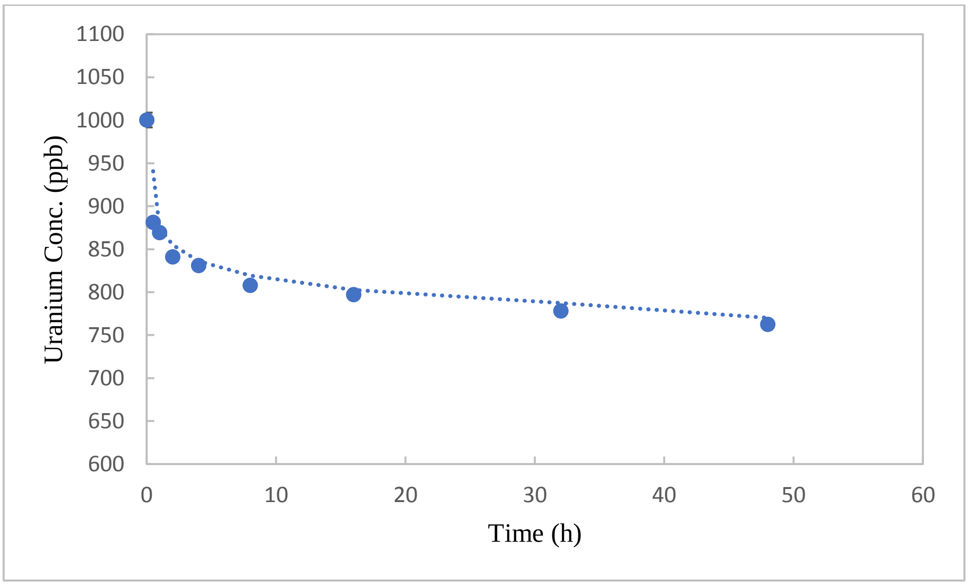

**Fig.2:**Kinetics of Uranium Adsorption onto Punjab Soil.

The adsorption isotherm indicates the solute distribution between the solid and liquid phase on reaching the adsorption equilibrium state (Hasan et al., 2008). On the basis of equilibrium time determined from kinetic study, batch isotherm experiments were performed for 48 hours at a pH of 7.4±0.1. The initial concentration of U(VI) was varied from 0.25 ppm to 1.25 ppm.

The difference between the initial U(VI)concentration and aqueous U(VI) concentration was attributed tothe adsorption of U(VI) onto the soil.The batch isotherm results are plotted in Fig. 3.

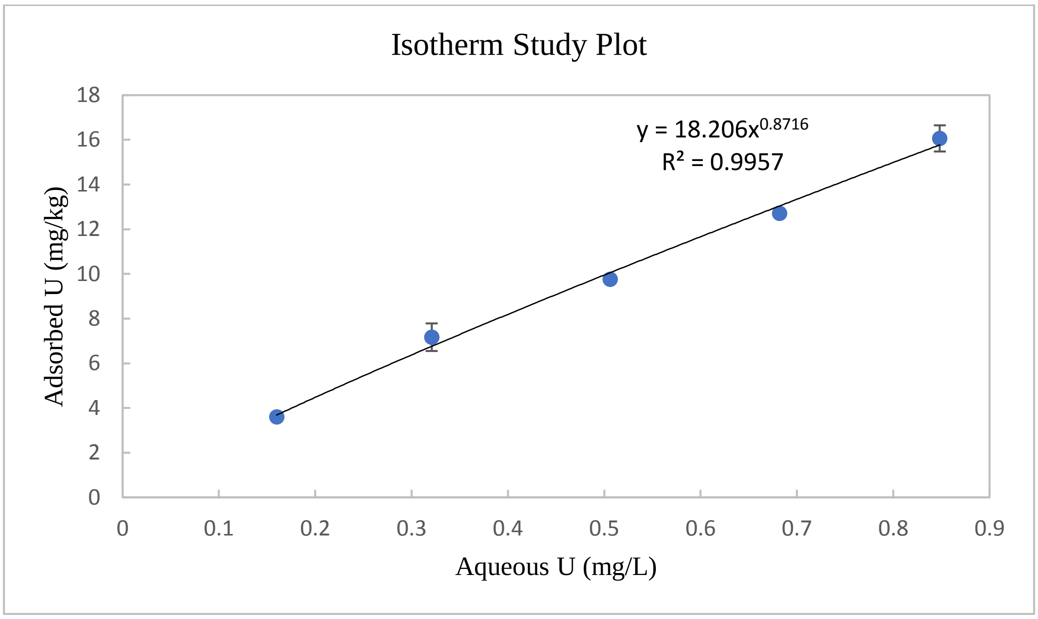

**Fig.3:**Adsorptionisotherm of Uranium onto Punjab Soil

The batch equilibrium isotherms are fitted by Freundlichequation ofthe form,

$$q_e = kC_e^n$$

kand $n$ are the Freundlich constants.

The pH of solution is one of the most importantparameters that control the adsorption of metal ions from aqueoussolutions (Kapoor et al., 1999; Zhang et al., 1998). The results of the pH studies are shown in Figure 4. The adsorption of U(VI) on to soils was maximum (36.48 percent of initial concentration) at pH of 7.02. At neutral pH, uranium adsorbs relatively strongly to mineral surfaces (Dittrich et al.,2015).

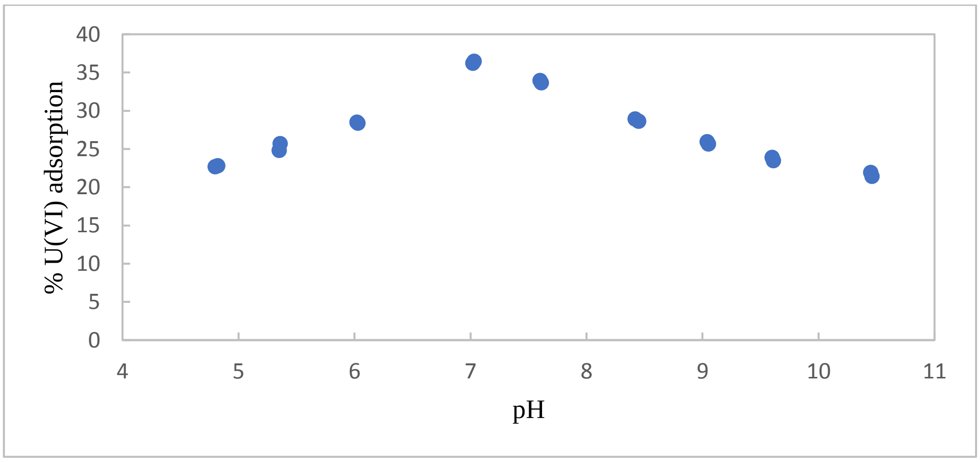

**Fig.4:** Effect of pH on adsorption of U(VI).

## 5. Conclusions

Our study was mainly aimed to decipher the rate of uranium adsorption onto the alluvial soil of Punjab and understanding the sorption processes under varied geochemical conditions that is representative of highly contaminated regions of Punjab. The results of the study indicate the following:

- The adsorption capacity of soil was mainly affected by the pH and reaction time. The adsorption of U(VI) increased with the increase of pH upto pH 7.0 and then decreases.
- Maximum adsorption of U(VI) was found to be 36.48% of initial concentration (1 ppm) and was found to be at near neutral condition.

These results will aid in better understanding of uranium mobility in the groundwater of Punjab region.

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## References

Bakatula, E.N., Mosai, A.K. and Tutu, H., 2015. Removal of uranium from aqueous solutions using ammonium-modified zeolite. *South African Journal of Chemistry*, *68*(1), pp.165-171.

Davis, J.A., Meece, D.E., Kohler, M. and Curtis, G.P., 2004. Approaches to surface complexationmodeling of uranium (VI) adsorption on aquifer sediments. *Geochimica et CosmochimicaActa*, *68*(18), pp.3621-3641.

Dittrich, T.M. and Reimus, P.W., 2015. Uranium transport in a crushed granodiorite: Experiments and reactive transport modeling. *Journal of contaminant hydrology*, *175*, pp.44-59.

Duff, M.C. and Amrhein, C., 1996. Uranium (VI) adsorption on goethite and soil in carbonate solutions. Soil Science Society of America Journal, 60(5), pp.1393-1400.

Fan, F.L., Qin, Z., Bai, J., Rong, W.D., Fan, F.Y., Tian, W., Wu, X.L., Wang, Y. and Zhao, L., 2012. Rapid removal of uranium from aqueous solutions using magnetic Fe3O4@ SiO2 composite particles. Journal of environmental radioactivity, 106, pp.40-46.

Hasan, M., Ahmad, A.L. and Hameed, B.H., 2008. Adsorption of reactive dye onto cross-linked chitosan/oil palm ash composite beads. Chemical Engineering Journal, 136(2-3), pp.164-172.

Kapoor, A., Viraraghavan, T. and Cullimore, D.R., 1999. Removal of heavy metals using the fungus Aspergillusniger. Bioresource technology, 70(1), pp.95-104.

Pabalan, R.T. and Turner, D.R., 1996. Uranium (6+) sorption on montmorillonite: Experimental and surface complexationmodeling study. Aquatic Geochemistry, 2(3), pp.203-226.

Patnaik, R., Lahiri, S., Chahar, V., Naskar, N., Sharma, P.K., Avhad, D.K., Bassan, M.K.T., Knolle, F., Schnug, E. and Srivastava, A., 2016. Study of uranium mobilization from Himalayan Siwaliks to the Malwa region of Punjab state in India. Journal of Radioanalytical and Nuclear Chemistry, 308(3), pp.913-918.

S. Bachmaf, P. Friedrich and B.J. Merkel, Effect of sulfate, carbonate, and phosphate on the uranium(VI) sorption behavior onto bentonite, *Radiochim. Acta*, 2008, **96**, 359–366.

Wan, J., Dong, W. and Tokunaga, T.K., 2011. Method to attenuate U (VI) mobility in acidic waste plumes using humic acids. Environmental science & technology, 45(6), pp.2331-2337.

World Health Organization, Uranium in drinking water. Background document for the development of WHO guidelines for drinking-water quality. WHO/SDE/WSH/03.04/118, 2004.

Zhang, L.I., Zhao, L.I., Yu, Y. and Chen, C., 1998. Removal of lead from aqueous solution by non-living Rhizopus nigricans. Water Research, 32(5), pp.1437-1444.