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GEOCHEMICAL MODELLING OF URANIUM SPECIATION AND ITS CORRELATION WITH SOME WATER QUALITY PARAMETERS OF WATER IN SOUTHEASTERN DISTRICT OF RAJASTHAN STATE, INDIA

[1]Ramet Meena, [2]Dhanraj, [3]Yogesh Kumar Tailor, [4]Ram Bilas Meena and [5]Suresh Kumar Meena
[1]Assistant Professor, Department of Chemistry, Govt. College, Kota, Rajasthan
[2]Assistant Professor, Department of Chemistry, Govt. College, Kota, Rajasthan
[3]Assistant Professor, Department of Chemistry, Govt. College, Kota, Rajasthan
[4]Assistant Professor, Department of Chemistry, Govt. College, Kota, Rajasthan
[5]Associate Professor, Department of Chemistry, Govt. College, Kota, Rajasthan

Abstract:
In the present study, analysis of uranium concentration in 20 drinking water samples collected from different locations of different division of southeastern district of Rajasthan state, India and LED fluorimeter technique was used to estimate the uranium content of water samples. The water samples were taken from hand pump, tube well, open well and dug well etc. The measured uranium concentration lies in the range from 0.9µg/l to 16.5$\mu g/l$ in pre mansoon with mean value of 3.822$\mu g/l$ and 1.1µg/l to 20.3µg/l in post mansoon with a mean value of 5.68µg/l. The measured uranium content in 20 water samples was found to be permissible limits range in the safe limit of 30µg/l as recommended by World Health Organization (WHO, 2011) and US Environmental Protection Agency (USEPA, 2011). An attempt has been made to correlate uranium concentration with these water quality parameters and a positive correlation of conductance, nitrate, chloride, TDS, calcium and hardness with uranium concentration has been observed and however no correlation of uranium concentration with pH was observed. Uranium associated with water quality parameters such as EC, pH, ORP, Phenolphthalein alkalinity, Temperature, Total Alkalinity, Total Hardness, Magnesium hardness, Calcium hardness, Chloride, Fluoride, Sulphate, Phosphate, Nitrate, Salinity, and Total dissolve solids (TDS) were also processed and analyzed using the BARC Standard Protocol.

Key words:LED Fluorimeter, Uranium, Water quality parameters, Southeastern district

Introduction

Groundwater is a vital resource, crucial for drinking, agriculture, and industrial purposes[1]. To ensure safety for human consumption, it must be free from harmful organisms and hazardous chemicals[2,3]. According to UNICEF's report "Fresh Water for India's Children and Nature," nearly 1 million children in India die annually from diarrheal diseases linked to unsafe water and poor sanitation[4]. Uranium, a dense, malleable, and weakly radioactive element, is naturally present in rocks, soil, air, and water[5-7]. It poses both chemical and radiological risks, particularly to the kidneys and lungs[8-10]. High concentrations of uranium in natural waters, typically above 100 µg/L, are rare and usually occur in aquifers with uranium mineralization[11-14]. Monitoring uranium levels in water serves dual purposes: it aids in the hydro geochemical exploration of uranium and assesses potential health risks[15-20]. Uranium concentrations in groundwater are influenced by local geology, including lithology and geomorphology[21]. This element is widespread in nature, particularly in granites and other mineral deposits, and its presence in drinking water is generally low[22-23]. However, when concentrations are elevated, the primary exposure route is through drinking water[24-27]. Natural uranium consists of three isotopes: 238U, 235U, and 233U. Of these, 235U is significant due to its ability to undergo fission, releasing substantial energy[28-33]. The hexavalent state of uranium is particularly important in water because of its solubility, unlike the tetravalent state, which is nearly insoluble[34-41]. Uranium can enter human tissues through various pathways, including ingestion, inhalation, and occupational exposure[42-45]. Factors affecting uranium concentrations in groundwater include pH, oxidation-reduction potential (ORP), electrical conductivity (EC), total dissolved solids (TDS), and anionic ligands[46-49]. The leaching of uranium from host rocks into groundwater is a complex process influenced by contact time, temperature, rock characteristics, and elemental geochemistry[50-54]. This study aims to assess the concentration of uranium and associated water quality parameters in drinking water from the Baran district of Rajasthan, India. Parameters analyzed include EC, TDS, pH, salinity, dissolved oxygen (DO), temperature, ORP, fluoride, chloride, nitrate, phosphate, sulfate, total hardness, calcium hardness, magnesium hardness, total alkalinity, phenolphthalein alkalinity, carbonate, and bicarbonate.

Materials and Methods

Geology of the Area

The geology of a region significantly influences groundwater occurrence and movement. In Baran district, the groundwater potential is dictated by the local geological formations. The area's basement comprises sandstone, limestone, and shale from the Bhander Group of the Vindhyan Supergroup, overlain by Deccan Trap basalt.

Estimation of Uranium and Water Quality Parameters

Uranium analysis was conducted using an LED fluorimeter (LF-2, Quantalase Enterprises Pvt. Ltd., India). The fluorimeter was calibrated with four uranium standards to ensure accurate performance and linear dynamic range. ORP, TDS, EC, pH, temperature, salinity, and DO were measured in situ using a waterproof portable meter (Eutech Instruments, Easy Cyberscan Series 600) with electrode sensors. Nitrate, chloride, and fluoride levels were also measured with the same portable meter. Total hardness and calcium hardness were determined by EDTA complexometric titration, with magnesium hardness calculated as the difference between total and calcium hardness. Total alkalinity was measured by titration with H2SO4 using methyl orange as an indicator. Phosphate and sulfate concentrations were analyzed using a UV-Visible Spectrophotometer (Lab India 3000+ UV/VIS).

Results and Discussion

In the southeastern district of Rajasthan, India, drinking water samples exhibited uranium concentrations ranging from 0.9 µg/L to 16.5 µg/L during the pre-monsoon period, with a mean value of 3.82 µg/L, and from 1.1 µg/L to 20.3 µg/L during the post-monsoon period, with a mean value of 5.68 µg/L. The U.S. Environmental Protection Agency (EPA) and the World Health Organization (WHO) have set a safe limit for uranium in public water supplies at 30 µg/L to minimize health risks over a lifetime of consumption. Understanding uranium speciation in these waters is crucial for predicting radionuclide migration and implementing effective water purification technologies. Uranium mining legacies and associated environmental hazards necessitate accurate thermodynamic data for affected environmental media. Thus, a comprehensive database was established to support ongoing and future assessments of uranium contamination and its impacts.According to current knowledgethe electrical conductivity of water of district ranges from 2.0-2677 µS/cm with average of electrical conductivity is 891.13 in pre monsoon & from 1.0- 2883 µS/cm with average of electrical conductivity is 838.85 in post mansoon. The pH value of water varies from 6.98 to 8.65 with average of pH is 7.53 in pre mansoon and 6.97 to 8.65 with average of pH is 7.53 in post mansoon. The reason for increase in TDS may be due to The minimum TDS value of the sample was found to be 2.0 ppm and maximum was found to be 2687 ppm, average of TDS is 897.91 In pre mansoon and minimum 1.0 ppm and maximum 2883 ppm, average of TDS is 838.85 in post mansoon. Calcium hardness value of water varies from 0.66-622 mg per litre, average of Ca hardness is 192 in pre mansoon and 40-650 mg per litre, average of Ca hardness is 180.81 in post mansoon. Mg hardness value of water varies from 0-440 mg per litre, average of Mg hardness is 119.62 in pre mansoon and 0-440 mg per litre, average of salinity is 111.84 in post mansoon. Correlation Analysis Correlation coefficient(r) is statistical measure of closeness of relationship between the independent variables and dependent variables.

Correlation Analysis

The direct correlation exists when; decrease or increase in value of one parameter is mainly associated with the corresponding increase or decrease in value of other parameters. The correlation is positive when increase in one of the parameter because an increase in other parameter and it is negative when increase in one parameter result in decrease of other parameter. Thus correlation coefficient (r) has values between +1 and -1. The correlation between parameters is categorized as strong, when it is in range of +0.8 to 1.0 and from -0.8 to -1.0, while it is weak when it falls in range from +0.0 to 0.5 and -0.0 to -0.5. **The highly competitive relationship**: - A correlation coefficient (r) of more than 0.5 indicated that the two variables were perfectly related in positive linear sense and correlated to each other parameter. In study area (i) during pre monsoon, correlation matrix depicts that Electrical Conductivity (EC) has positive correlation with Total Dissolved Solids (TDS), where correlation coefficient, (r) is (+ 0.99). TDS describes solids which are dissolved in water. The more salts dissolved in water, the higher will be the value of Electrical Conductivity. In groundwater samples during post monsoon TDS with EC (+0.96) positive correlation shows the presence of some inorganic and organic salts in the drinking water. (ii) A significant correlation coefficient (r) exists between Total-Hardness and Ca-Hardness (+0.90), Mg-Hardness (0.80) during pre monsoon and in ground water samples during post-mansoon Total Hardness with Ca-Hardness (+0.91), Mg-Hardness (+0.79) positive correlation shows the presence of some weathering of magnesium bearing minerals, agricultural runoff and mixing of industrial effluents. (iii) thus strong positive correlation coefficient exist between Phenolphthalein Alkalinity and Carbonate (+0.97) in during the pre monsoon season and in post monsoon, the correlation coefficient is (+0.965). (iv) a significant correlation coefficient (r) exists between Total alkalinity and Bicarbonate (+0.79) during pre monsoon ,in the Ground water samples during post mansoon Total alkalinity with Bicarbonate (+0.76) strong positive correlation. (v) A significant correlation coefficient (r) exists between Total Dissolve Solids (TDS) and Salinity (+0.53) during post- monsoon. These types of significant positive correlation between the ions indicated that the ions are from the same source of origin. **The affinity ion relationship**: - A correlation coefficient of less than 0.5 indicates that two variables are not significantly correlated to each other as there is linear relationship between the various variables. **The non-competitive relationship**: - The correlation coefficient of -1 indicated that the two variables were not correlated each other. The correlation analysis of water quality parameters reported that parameters are more or less correlated with each other. It is observed that some of the parameters do not have significant correlation between them indicating the different origin of source of pollution. The correlation matrix was prepared to check relation between the different groundwater quality parameters of both pre monsoon and post monsoon season which are presented below in Table 1 respectively.

Table 1 Correlation coefficients and the nature of correlation for uranium measurements with different water quality parameters in drinking water samples in pre-monsoon and post monsoon

S.No.	Water quality parameters	Correlation coefficient (r) in pre monsoon	Nature of correlation in pre monsoon	Correlation coefficient (r) in post monsoon	Nature of correlation in post monsoon
1	pH	-0.00	No correlation	0.04	Weak positive correlation
2	TDS	0.04	Weak positive correlation	0.05	Weak positive correlation
3	EC	0.04	Weak positive correlation	0.05	Weak positive correlation
4	ORP	-0.05	Weak negative correlation	-0.05	Weak negative correlation
5	TEMP.	0.01	Weak negative correlation	-0.18	Weak negative
6	Salinity	-0.09	Weak negative correlation	-0.00	No correlation
7	DO	-0.13	Weak negative correlation	-0.17	Weak negative
8	Fluoride	0.32	Weak positive	0.32	Weak positive
9	Chloride	-0.05	Weak negative correlation	-0.02	Weak negative correlation
10	Nitrate	0.07	Weak positive correlation	0.06	Weak positive correlation
11	Sulphate	0.06	Weak positive correlation	0.06	Weak positive correlation
12	Phosphate	-0.07	Weak negative correlation	0.01	Weak positive correlation
13	Total-Hardness	0.18	Weak positive	0.19	Weak positive
14	Ca-Hardness	0.10	Weak positive	0.11	Weak positive
15	Mg-Hardness	0.25	Weak positive	0.25	Weak positive
16	Phenolphthalein alkanity	0.03	Weak positive correlation	0.04	Weak positive correlation
17	Total alkanity	0.03	Weak positive correlation	0.07	Weak positive correlation
18	Carbonate	0.01	Weak positive correlation	0.04	Weak positive correlation
19	Bicarbonate	0.01	Weak positive correlation	0.02	Weak positive correlation

Conclusions:

Considering the aforementioned analytical capabilities, the observed distribution of uranyl speciation in neutral and alkaline model systems and comparable waters, such as shallow groundwater and pore water, is reasonably well-represented. However, further investigations are needed for waters with significantly different compositions, such as acidic or sulfate-dominated waters. Common thermodynamic databases used in geochemical modeling software vary greatly in completeness, consistency, and recent knowledge regarding uranium speciation in aquatic environments. This is particularly significant for stable earth alkali uranyl carbonate and low-solubility phosphate uranyl complexes, which are crucial for understanding uranium migration in neutral and alkaline systems. This paper aims to enhance the sensitivity of geochemical modelers to this fundamental issue and seeks to optimize the thermodynamic database while evaluating its accuracy for various natural high-uranium aquatic systems. The uranium levels in several drinking water samples from the southeastern district exceed safety limits, rendering them unsafe for consumption. The study concludes that while uranium concentrations in the district are within permissible limits for groundwater used for drinking, other parameters such as Total Hardness, Total Dissolved Solids (TDS), Salinity, Alkalinity, Fluoride, and Phosphate often exceed permissible limits. Post-monsoon averages for salinity, fluoride, nitrate, total alkalinity, phosphate, bicarbonate, and uranium are higher than pre-monsoon averages, reflecting the potential leaching of salts during the rainy season, which increases their correlation in groundwater. Regular quantitative and qualitative measurements are essential to continuously monitor physicochemical water quality parameters from various groundwater sources and to implement appropriate remediation strategies. Uranium shows a weak positive correlation with Total Hardness, Calcium Hardness, Magnesium Hardness, Total alkalinity, Bicarbonate, Carbonate, and Phenolphthalein alkalinity during the pre-monsoon and with Carbonate, Bicarbonate, Phosphate, Phenolphthalein alkalinity, Total alkalinity, Total Hardness, Calcium Hardness, and Magnesium Hardness during the post-monsoon. However, there is no correlation between uranium concentration and pH, Oxidation-Reduction Potential (ORP), Dissolved Oxygen (DO), Chloride, and Phosphate in the pre-monsoon or with ORP, Temperature, Salinity, and Chloride in the post-monsoon. The elevated uranium levels suggest that water in these regions is unsafe for drinking and poses health risks.

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