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Hydrochemistry and Uranium Concentration in Brackish Groundwater from an Arid Zone, Chihuahua, Mexico

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ABSTRACT

In arid zones, the principal water supply is from groundwater, which can present low-quality for its use due to the high concentrations of salts, heavy metals, and radioactive elements. The aim of the study was to determine isotopic uranium concentration in groundwater samples with a high concentration of salts and their association with other chemical species. Groundwater samples were taken from wells that shows high salt contents. The ^{238,234}U radioisotope concentrations were determined by liquid scintillation and alpha-particle spectrometry. In addition, the physical-chemical parameters were recorded in situ; whereas the dissolved ions and elemental composition were measured using a UV-Vis spectrophotometer and X-ray fluorescence spectrophotometry, respectively. To obtain isotopic uranium concentrations, three radiochemistry procedures were carried out. An ANOVA test was performed to compare the results from procedures, as well as an analysis of the Pearson correlation was used to obtain their associations. Statistically, the U isotopic concentrations did not show differences (0.82 p<0.5) between procedures. ²³⁸U and ²³⁴U showed mean concentrations of 6.7 mBq mL⁻¹ and 16.6 mBq mL⁻¹, respectively, with an Activity Ratio by up 7.2. The groundwater under study showed high concentration of TDS, calcium, sulphate, chloride, nitrate, and nitrite. Isotopic U concentrations tend to increase with NO₃>Zn>Cl>Br>SO₄>Cu>T>SDT>P; meanwhile, their contents decrease with T>Cl>NO₂>Fe. These findings help to understand the uranium behavior in groundwater with high salt contents as well as the influence of agricultural supplies on chemical species presents in groundwater.

1. Introduction

Arid zones are those with mean annual precipitation less than 350 mm [1]. These areas are characterized by water scarcity, erratic, and torrential distribution of rainfall, as well as the intensive use of water for economic activities. Thus, in these regions, groundwater is the main water supply for several natural and anthropogenic activities. Therefore, the overexploitation of aquifers is a current problem that leads to the depletion of global water resources [2]. In arid regions, groundwater tends to have high content of dissolved salts, which represents a disadvantage for human use and consumption.

Uranium (U) is a natural radioactive element with three isotopes: 238 U (99.2739%), 235 U (0.7205%), and 234 U (0.0056%). In nature, U is found as salts and oxides

showing different compositions [3, 4]. The main effect of U over environment is its compounds toxicity. The U geochemical cycle begins with the weathering from the earth's crust leading to U mobilization in surface and ground water, through particles or dissolved fractions [5]. The oxidation-reduction conditions plays an essential role in the U mobilization; under reduction conditions, U is mainly in + 4 oxidation state and is insoluble while in oxidation conditions it takes + 6 oxidation state and forms soluble ions (OH⁻, CO₃⁻², F⁻, SO₄⁻², PO₄⁻³)[6, 7].

The detection of U in water samples have been carried out with non-radiometric techniques (e.g. ICP-MS, atomic absorption and fluorescence spectroscopy) and radiometric techniques (e.g. liquid scintillation, alphaparticle spectrometry). The first one consists in total U determination without isotopic information meanwhile the second one gives information about active radios of U isotopes [8].

Previous results demonstrated the presence of U in shallow and groundwater samples in Chihuahua, Mexico by radiometric techniques [9]. However, radiometric techniques are limited by high element/salt concentrations resulting in low-quality spectrum and yields. Authors have proposed some improvements to the current procedures, these are apply pre-concentration, separation of radionuclides by co-precipitation, and ion exchange resin [10, 11]. Therefore, it is important to obtain methodologies which lead the radioactive measurement of samples with high salt concentration as in groundwater from arid zones. This study aimed to determine ^{238,234}U concentrations in groundwater samples and their geochemical relationship with other chemical species. These results will be a baseline for this type of study and will provide help in water treatment studies.

2. Materials and Methods

Study Site

The aquifer of Jimenez-Camargo is located in the extreme southeast of the state of Chihuahua with an area of 9947.7 km². It is located between the coordinates 27°08" North and 104°55" West, at a height of 1,380 m above sea level. The climate is semiarid, with a mean annual temperature of 18.66 °C and an average annual precipitation of 380 mm [12]. The surface of the aquifer is almost completely

composed of alluvial material located in the lower parts from the north to the southeast. It is an unconfined aquifer with some semiconfined zones. The surface is composed of sand and gravel, and can even contain accumulated clay and/or carbonate-sulphate minerals; therefore, its nature generates a high content of some elements [13]. The soil of the aquifer is mainly composed of xerosol which is characteristic of dry areas, with a clear and thin surface, with highly variable amounts of organic matter depending on the type of texture. Under this layer, there is accumulation of clay minerals and/or carbonates/sulfates; at a certain depth there are spots, lime agglomerations, and gypsum crystals with different levels of salts [14], Pb, Fe, Zn, Mn & Cu [15, 16]. Figure 1 shows the location of the study area, as well as the sampling points.

Water Sampling

The groundwater from 30 wells, which are used for irrigation of pecan-producing orchards, was analyzed. Samples were taken in two types of containers: a) sterile polypropylene containers of 1 L [17] for determination of dissolved ions (which were preserved on ice to $4^{\circ}C \pm 1^{\circ}C$ and b) containers of 15 L for determination of isotopic U, as well as major and trace elements. Sample locations were determined randomly; these sampling points were located using a global positioning system (GPS). Parameters such as temperature (T), pH, turbidity (TB), and total dissolved solids (TDS), were measured in situ using multi-parametric equipment (Hanna, HI98130) and a portable turbidimeter (Hanna, HI, 93703).



Figure 1: Location of the sampled wells.

2.1. Uranium Isotopic Determination

Radiometric techniques are limited by high element/salt concentrations resulting in low-quality spectrum and yields. Due to this, in radiometric methodologies, it is necessary to improve the isolation of the main element previous to its measurement. In this study, three radiometric procedures were applied to determine ^{238,234}U concentrations in groundwater with high salt concentration from 7 wells. Prior to radiochemical treatments, 3L of sample water aliquots was concentrated to 250 mL by evaporation; treatments applied were: 1) actinide extraction using tributyl phosphate (TBP) [18], which is widely used to analyze U in environmental samples using semiconductor detector PIPS, 2) the previous procedure to which a filter step is added, and 3) URAEX-extract procedure under sulfate media [19, 20] using liquid scintillation (PERALS, Photon-Electron Rejecting Alpha Liquid Scintillation).

Procedure One (P1)

50 mL of pre-concentrated samples were put under analysis; each aliquot was spiked with 0.5 mL of ²³²U (98.12 mBq mL⁻¹) for traceability. The actinide precipitation was carried out with Fe⁺³ and, after this, the precipitate was separated from the liquid phase by centrifugation (Hettich, Rotofix 32A). ^{238,234}U extraction took place with TBP (tributyl phosphate, SIGMA-ALDRICH). Then, uranium isotopic isolation was performed by electrode position method, to 1.2A during one hour. Finally, an alpha-spectrometer (Alpha analyst, CANBERRA) was used for isotopic U measurements. The uranium activity concentrations were calculated by equation (1) proposed by Kumar et al. [21].

$$A(mBqL^{-1}) = \frac{Cps\alpha_m}{R\% \times V} \tag{1}$$

where $CPS\alpha$ is counts per second in the alpha region, R is chemical recovery and V is sample volume.

Procedure Two (P2)

50 mL of pre-concentrated samples were filtrated by a 0.45 μ m membrane (EMD Millipore S-Pak, HAWG047S6). Then the sample followed the same treatment that procedure one: tracer addition, actinide precipitation with Fe⁺³ solution, U extraction with TBP, electrode position and alpha-spectrometry counting. Isotopic U was calculated by equation (1).

Procedure Three (P3)

50 mL of pre-concentrated samples were diluted 1:4 with distilled water, reaching 250 mL of total volume. To this aliquot, 0.3 gr of ²³²U (High Purity Standards No. 100064-

1; 128.88 mBq L^{-1}) were added as a tracer, then, this solution was carried out to dryness. This procedure was according to McKlveen, J.W. and W.J. McDowell [19]. The activity of U was counting by PERALS spectrometer (Oak Ridge TN, EUA). Isotopic U was calculated by equation (1).

2.2. Measure Techniques for Chemical Species in Groundwater

To determine anions and cations a UV-Vis HACH d3900 spectrophotometer was used; meanwhile the determination of the element composition was done with TXRF, using a Bruker S2 PICOFOX. Both techniques are described in [22].

2.3. Statistical Analysis

To determine the origin of these variables, a Pearson correlation was carried out using the procedure CORR in the SAS package, to P < 0.05 and P < 0.01. Likewise, an ANOVA test was carried out for the three procedure comparison using procedure PROC GLM [23], to P < 0.05.

3. Results and Discussion

3.1. Radiochemistry Characterization

Water monitoring is essential to public health, automating routine as physical-chemical parameters, microbiology, among others. The high content of salts in a sample inhibits the direct extraction of certain chemical species such as radionuclides [24]. The detection of those elements by alpha emission requires a procedure with high extraction reliability. Thus, the insolation of these alpha emitters requires an adequate radiochemistry methodology, followed by the electrode position process and alpha spectrometry counting.

Isotopic U activity concentrations obtained from every procedure are shown in Table 1. The averages of activity concentration were 5, 7, and 9 mBq mL⁻¹ of ²³⁸U in P1, P2, and P3, respectively. Whereas, ²³⁴U activity concentrations were 16, 20, and 17 mBq mL⁻¹ in P1, P2, and P3, respectively. The activity ratio (AR, ²³⁴U/²³⁸U) for the three procedures were greater than one, in an interval of 1.2 to 7.2.

Moreover, the ANOVA test showed that results obtained from procedures were similar statistically (p-value < 0.5). It is known high salt concentration in water difficulty the U detection [11]. For actinide separation is common to use different types of resins and solvents [25, 26]. However, DOWEX 1-8X and UTEVA resins have shown interferences on the insolation of U for these water samples [20]. For P1 and P2, activity concentrations of isotopic uranium were similar, however, the filtering step used in P2 improved the measurement yields. This result is similar to that found in seawater analyzed [27]. Likewise, the groundwater samples analyzed by liquid scintillation (P3) showed similar yields as those obtained from alpha spectrometry. Authors have been compared some methodologies to obtain U activity concentrations, and they are found better results by alpha spectrometry than those measured by liquid scintillation [18]. Aliquots volume was not interference in the modification of procedures; due to all sample volumes used were of 3L [25, 28].

On the other hand, U activity concentrations were above to the recommended limit for gross alpha emitters in drinking water of (0.56 Bq/L) by Mexican regulations [29], as well as the recommended limit to ²³⁸U contents in drinking water of 0.37 Bq/L by EPA [30]. In a previous study done in groundwater samples from Jimenez-Camargo aquifer, Ut activity concentrations were up to 0.266 mBq mL⁻¹; being below to the results of this study [31]. This increase of Ut activity concentration may be attributed to several causes, some of those can be related to U leaching (under oxidation conditions) from aquifer material containing high U concentrations such as clay lenses and/or rocks [32]. In this region, a uranium deposit is placed to the southwest [33] which can be the main contributor of natural radioisotopes to groundwater. Moreover, this phenomenon can be exacerbated by the groundwater overexploitation, due to this water is abstracted from deeper aquifer places where dissolved material is in higher concentrations [13, 18]. Likewise, concentrations of Ut obtained in groundwater under analysis were higher than those found in aquifers with similar geological and climatic conditions [32, 34, 35]. Furthermore, AR indicated a preferential dissolution of ²³⁴U over ²³⁸U, generating a radioactive disequilibrium. This is the result of geochemical differentiation processes; in which the decay chain daughter (²³⁴U) is more mobile than the father (²³⁸U). In this process alpha energy causes recoil fragmentation between radionuclides ²³⁴U and ²³⁸U; mobilizing ²³⁴U from the surface of water rocks resulting in disequilibrium in the solid and liquid phase [36]. Thus, AR helps to understand the hydrological and geochemical processes in which can be groundwater markers [37, 38]. Therefore, low concentrations of ²³⁸U and higher RA values can suggest a deeper source of water in this aquifer [39], as well as a possible water mix with groundwater from thermal origin [11].

	238 U	$^{234}\mathbf{U}$		Ut	Yield
	$\mathbf{mBq} \ \mathbf{mL}^{-1}$	$mBq mL^{-1}$	AR	$mBq mL^{-1}$	(%)
		Procedu	ure one (P1)		
Minimum	1.00	2.40	1.81	3.40	34.91
Maximum	9.52	31.86	5.39	41.38	77
Mean	5.13	15.87	3.12	21.00	55.82
SD	3.07	11.00	1.32	13.75	15.95
CV	59.90	69.34	42.37	65.49	28.58
		Procedure	e two (P2)		
Minimum	1.95	4	1.67	6.4	16.27
Maximum	20	49.2	7.24	69.16	97
Mean	6.98	20.39	3.37	27.37	41.8
SD	7.01	17.65	2.07	24.41	33.1
CV	100.41	86.57	61.33	89.21	74.24
		Procedure	three (P3)		
Minimum	4.81	10.5	1.19	16.24	36.26
Maximum	13.32	23.41	2.65	33.91	86.57
Mean	9.14	16.48	1.89	25.62	64.36
SD	3.28	5.59	0.50	8.18	18.19
CV	35.87	33.94	26.75	31.94	28.27

Table 1: Activity concentration of ²³⁴U, ²³⁸U, total U (mBq mL⁻¹), as well as the activity relationship (²³⁴U/²³⁸U) and yield (%) in salty water samples.

*uncertainty <30%; ** uncertainty <20%; SD, Standard deviation; CV, Coefficient of variation

3.2. Elemental Correlations

The result of trace elements and dissolve ions concentrations for these wells were published in

Table 2: Uranium correlations with water quality variables.

[22]. Significant correlations for U are showed in Table 2. Thus, U showed positive correlations were for $NO_3>Zn>Cl>Br>SO_4>Cu>Tb>SDT>P$; meanwhile negative correlations were for $T>Cl->NO_3>Fe$.

	TB	Т	TDS	Cl-	SO_4	NO ₃	NO ₂	Р	Cl	Fe	Cu	Zn	Br
²³⁸ U	0.66*	-0.82^{*}	0.43*	-0.50^{*}	0.68*	0.94*	-0.50^{*}	0.35	0.74*	-0.35	0.69*	0.92*	0.73*
²³⁴ U	0.56*	-0.76^{*}	0.46*	-0.44^{*}	0.73*	0.92*	-0.48^{*}	0.42*	0.78*	-0.42^{*}	0.67*	0.89*	0.77*
Ut	0.64*	-0.81^{*}	0.44*	-0.49^{*}	0.70*	0.94*	-0.50^{*}	0.37	0.75*	-0.37	0.69*	0.92*	0.74*
	0.01	0.01	0.11	0.17	0.70	0.71	0.90	0.57	0.79	0.57	0.07	0.72	0.7 1

* Significant level 0.05

Water chemistry plays an important role in U complex formation with different ligands. Also, 5% of the recognized minerals have U, showing a wide diversity in the behavior of this element [40]. In semiarid conditions under high oxidations water increase the U solubility [41]. According to results, U is highly correlated with NO₂ (0.93 p<0.05); this is related to agricultural activities [42]. In high oxygen environment, the nitrates concentrations are significant. Moreover, it had been found that nitrates presence may increase U mobility [43]. Furthermore, excessive irrigation and agricultural practices have been the main reasons of U leaching in groundwater. Additionally, U showed positive correlations with cations such as Zn (0.91 p<0.5) and Cu (0.68 p<0.5), due to all of them can be complexed with sulfates. In agriculture practices, zinc sulfate is widely used as a foliar additive. Due to dissolution Zn may release from that molecule and U could bonded with sulphate [20]. It can explain the U correlation with SO₄ (0.70 p < 0.5)which can be from agricultural or geological sources from the youngest sediment layers [8, 44, 45]. This is similar to some studies where high U concentrations in groundwater were found from quaternary and tertiary sediments regions that exhibited U detectable concentrations meanwhile in paozoic and Jurassic rock sediments show low detectable concentration [46]. Likewise, Cu had a geological origin by sulfate minerals that facilitate U complex, releasing Cu [47]. Groundwater from this zone was classified as calcic-sulfated due to the calcite rocks in the zone [13]. Also, sulfate, Ca, and Mg concentration increase by evaporites dissolutions reflected in dissolve ions by parameters like TDS [48]. In addition, U also showed correlation with TB (0.62 p<0.5) and TDS (0.44 p < 0.5), indicating a geological complex zone. U showed a positive correlation with Cl (0.76 p < 0.5)and Br (0.75 p<0.5) from minerals, corroborating zones with high dissolved ions [40]. Finally, P (0.38 p<0.5) can be associated with phosphate fertilizers that increase U concentration [49].

On the other hand, U shows a negative correlation with T (-0.80 p < 0.5). A previous study show similarity

in groundwater [41]; corroborating the influence of T to kinetic reactions for different U complex (mainly with carbonates) more than its distribution [32]. Likewise, U concentrations decrease in presence of Cl⁻ (-0.48 p < 0.5) and NO₂ concentration (-0.5 p < 0.5); it can suggest the presence of reduced zones in which U takes +4 oxidation state and is precipitated [5]. Finally, U showed a significate negative correlation with Fe (-0.38 p < 0.5) which is attributed to the dissolution and formation of some U complex [7].

Conclusions

In this study, isotopic uranium concentration was obtained from salty groundwater samples. From a statistical point of view, uranium contents from groundwater analyzed by the three procedures were similar. Furthermore, the isotopic uranium concentrations in groundwater under study are attributed to a geological source, from deeper aquifer zones. The agricultural practices in that region can be increasing the isotopic uranium concentrations due to the intensive use of water for irrigation.

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