

## Radioactive hydrogeochemical processes in the Chihuahua-Sacramento Basin, Mexico

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The Chihuahua Basin is divided by its morphology into three main subbasins: Chihuahua-Sacramento subbasin, Chihuahua Dam subbasin and Chuvíscar River subbasin. In the aquifers at the Sacramento subbasin, specific concentrations of uranium in groundwater range from 460 to 1260 Bq / m<sup>3</sup>. The presence of strata and sandy clay lenses with radiometric anomalies in the NW of Chihuahua Valley was confirmed by a lithostatigraphic study and gamma spectrometry measurements of drill cuttings. High uranium activity values found in the water of some deep wells may correspond to the presence of fine material bodies of carbonaceous material, possible forming paleo-sediment of flooding or paleo-soils. It is suggested that these clay horizons are uranyl ion collectors. Uranyl may suffer a reduction process by organic material. Furthermore the groundwater, depending on its pH and Eh, oxidizes and re-dissolves uranium. The hydrogeochemical behavior of San Marcos dam and the NW Valley area is the subject of studies that should help to clarify the origin of the radioactive elements and their relationships with other pollutants in the watershed.

*Keywords:* uranium; groundwater quality; sediments.

La cuenca hidrológica de Chihuahua se divide por su geomorfología en tres principales subcuencas: Subcuenca Chihuahua-Sacramento, Subcuenca de la presa Chihuahua y Subcuenca del río Chuvíscar. En los acuíferos de la subcuenca Sacramento, las concentraciones específicas del uranio en agua subterránea fluctúan de 460 a 1260 Bq/m<sup>3</sup>. Se comprobó la localización de estratos y lentes arcillo-arenosos con anomalías radiométricas en la zona NW del valle Chihuahua, mediante un estudio litoestadigráfico de barrenos y por mediciones de espectrometría gamma. Las anomalías encontradas en el agua de algunos pozos profundos se corresponden con la presencia de cuerpos de material fino areno-arcillosos mezclado con material carbonoso, formando posibles paleo-suelos o paleo-sedimentos de inundación. Se sugiere que estos horizontes areno-arcillosos son recolectores del ion uranilo y hacen que el uranilo sufra un proceso de reducción por el material orgánico. Después el agua subterránea, en dependencia del pH y el Eh, oxida nuevamente y redissuelve el uranio, pasando al agua que se extrae de los pozos. La hidrogeoquímica de la presa de San Marcos y de la zona NW del valle es objeto de estudios que deben contribuir a esclarecer el origen de los elementos radiactivos y sus relaciones con otros contaminantes en la cuenca.

*Descriptores:* Uranio; calidad del agua subterránea; sedimentos.

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### 1. Introduction

The first specific studies on radioactive contamination of drinking water in the city of Chihuahua started with the investigation of radioactivity in sediments, surface and well waters in different locations at the basin and the Chihuahua State [1-8].

The hydrological basin of Chihuahua is divided by its geomorphology into three main basins [9]: Chihuahua-Sacramento Subbasin, Chihuahua Dam Subbasin and Chuvíscar River Subbasin. To give the geological context, in Figure 1 a satellite image showing the tectonic of the Chihuahua Valley zone is presented. In the figure are shown the main streams of Sacramento River, which results from Majalca and San Marcos streams and then it runs at the East of Chihuahua Valley. The streams forming the Chuvíscar River, which runs at South of the valley are also shown. Sacramento River joints to Chuvíscar River, which continues to the North-East, not represented in Figure 1.

In the aquifers of the Chihuahua-Sacramento subbasin, specific concentrations of uranium in groundwater range from 0.46 to 1.22 Bq / L of water [1]. In that study 29 wells that supply the city of Chihuahua were analyzed and up to 80% had activity concentrations of uranium or radium above the standard (0.56 Bq / L) [10]. These elements dissolved in the water are a sign of the possible presence of uranium in the sediments of the alluvial filling. The source of Sacramento River is in the vicinity of a major uranium deposit, which suggests being the origin of radioactivity downstream, i.e., uranium concentrations found in water from wells at Chihuahua-Sacramento Valley.

To clarify the cause of the contamination described above was conducted a characterization work of the reservoir [11] and of the uranium-series isotopes content in sediments, surface water, groundwater, fish and plants in the area of San Marcos, one of the origins of the Sacramento River in Chihuahua [12-15].

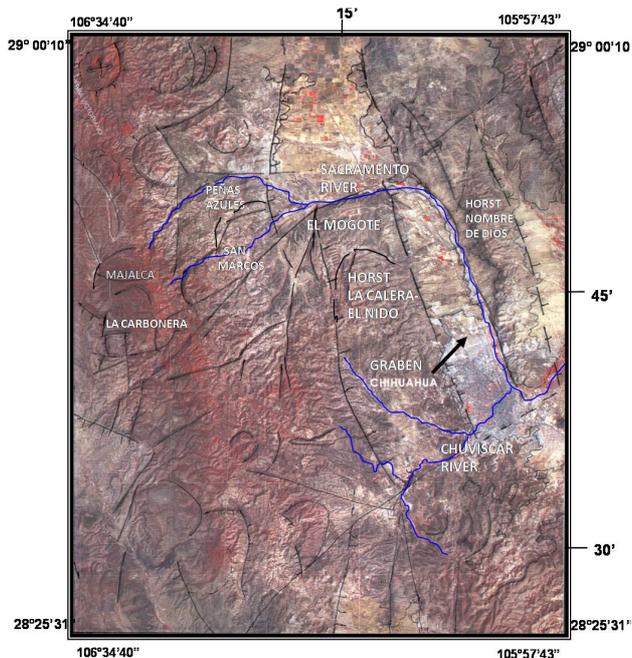


FIGURE 1. Tectonic of Chihuahua City area, showing horst and graben types of blocks. Rivers from Chihuahua-Sacramento and Chihuahua Dam subbasins are also shown.

In this paper were performed calculations for obtaining concentrations trends of total uranium activity in the Chihuahua-Sacramento River subbasin. Also was performed a petrographic and mineralogical study of powder and splinters materials from drilling of deep wells in the valley, provided by the Central Board of Water and Drainage (JCAS). Moreover, the uranium specific activity of this material was measured and correlated with the presence of lacustrine material. All this analysis contributes to the development of a conceptual model to explain the uranium activity in water from wells at the basin [16]. The current work contributes to the better explanation of different phenomena.

## 2. Materials and methods

### 2.1. Groundwater radioactivity study

In 2006, a sampling campaign of 24 groundwater wells in the Chihuahua-Sacramento Valley was performed. Samples of 5 liters in volume were acidified to pH 2 with nitric acid. They were analyzed by the method of uranium extraction from water with bis (2-ethylhexyl) phosphoric acid (HDEHP) in a separating funnel. The organic phase was completed up to 20 mL with Ultima Gold AB liquid scintillator [17]. Then samples were measured in a liquid scintillation detector with alpha-beta separation Triathler OY. Through the comparison with an external standard, the total activity of uranium isotopes in the sample was assessed.

With the results of activity concentrations was performed an interpolation by drawing isoconcentration lines in the plane of the subbasin and a representation in space, where the dependent variable is the specific activity. On calculations

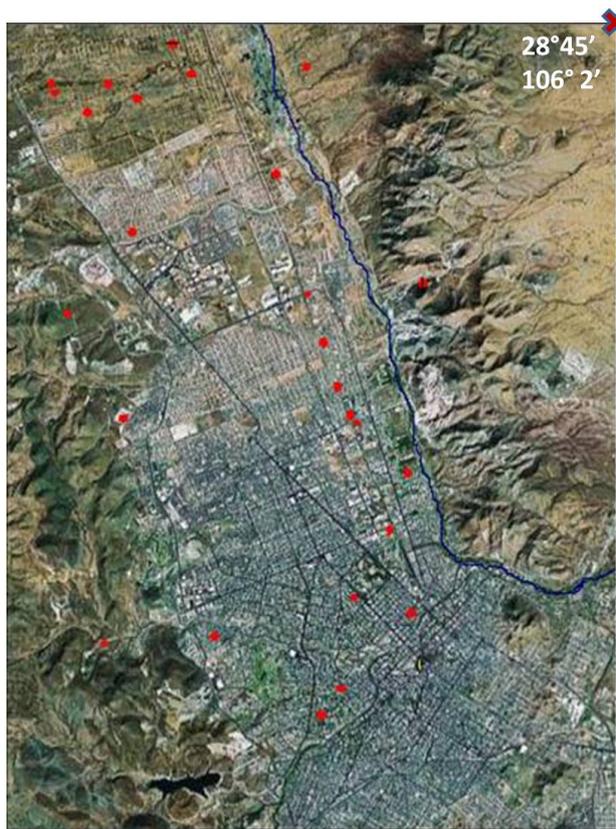


FIGURE 2. Location of groundwater wells considered in calculations.

were added the activity concentrations obtained in a sampling carried out in 2003 and published in [1] of 11 wells at the basin, with the repetition of 6 wells in the sampling of 2006. Repeated data were averaged. A total of 29 wells were studied, of different ages and drilling depths.

Figure 2 shows the position of 26 wells considered in the two sampling campaigns. The wells very close, as are the 3 adjacent to the Cerro Picacho, are shown as one and the average activity concentration obtained in all three was introduced into calculations.

### 2.2. Aquifer host rocks radioactivity study

For the radioactivity investigation in the aquifer strata, were studied samples of drilling fragments and powder, provided by JCAS and obtained from private wells. JCAS water wells generally vary between 200 and 500 m deep. Samples of fragments and powder were recovered every 2 m. More than 1750 samples from these boreholes were observed under binocular microscope to determine qualitatively particle size and megascopic characteristics. The samples at first instance were classified as fresh rock, alluvial conglomerate and breccia, sandy material and clay material. These materials were carefully observed to determine their possible origin as organic soil or lake sediment. The main objective was to determine the possible lake horizons or paleo-soils and to compare them with results of radiometric analysis. It was done to verify



FIGURE 3. Classification of fragments and powder samples extracted from RS6 well: to the left is the appearance of clay lake bottom material; at center, sandy alluvial material; at right, fragments of ignimbritic rhyolitic rocks.

if really sandy or clay horizons in the subsoil were able to capture the uranyl ion formed by U (VI) to precipitate it as U (IV). This capture has been proposed on the base presented by [18]. Figure 3 presents the gross aspect of different kind of samples extracted from the RS 6 well. For purposes of this research was also obtained information from electric logs made during drilling, which allows to deduce the lithology of the strata that cross wells.

From the large body of samples were selected 220 in sections of 2 to 10 meters and prepared for radiometric study. From each sample were separated fraction of grains smaller than 2 mm in diameter (hereafter called fine), and recorded the mass of both the coarse fraction as of the fine. The specific activities of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the fine fractions were determined by gamma spectrometry (GS). A CANBERRA spectrometer was used based on a  $10 \times 10$  cm NaI(Tl) detector with the program GENIE 2000. To implement the calculation for long times were measured four spectra of Standard Reference Materials IAEA RGK-1 ( $^{40}\text{K}$ ) RGTh-1 ( $^{232}\text{Th}$ ) and RGU-1 ( $^{238}\text{U}$ ) and laboratory background radiation. For data reduction of spectra the Standard Striping program was used, carried out by [19]. The program is based on the calculation of the contribution of each reference spectrum to the spectrum of the study sample [20]. In preparing the program was applied the Monte Carlo method for calculating the detection efficiencies in the geometry of the experiment [21-24]. The results of the  $^{238}\text{U}$  activity of the fine fraction were divided by the total mass of the sample and recorded as  $\text{U}/m_{tot}$  in units of  $\text{Bq} / \text{kg}$ .

### 3. Results

#### 3.1. Groundwater radioactivity study

Table I shows the total uranium specific activities from groundwater reported in [1], and Table II, the same data obtained from the sampling in this work. An analysis of statistical distribution of the specific activities of the whole set was carried out, proving its lognormal character. It is said that the lognormal character of a distribution of abnormally high con-

centrations of a substance in an area attests the multifactorial origin of the phenomenon. An example of the application of the lognormal distribution to anomalous values of natural radioactivity can be found in [25].

The specific activities values of uranium in groundwater range from 0.28 to 1.60  $\text{Bq} / \text{L}$ . Of 29 wells tested that supply drinking water, 45% contain uranium concentrations above the Mexican norm (0.56  $\text{Bq} / \text{L}$ ) [10]. These trace elements dissolved in the water are a sign of the possible presence of uranium in the sediments of the alluvial filling. In parallel experiments, the presence of radium has not been always detected.

A model based on an interpolation of the total specific activities of uranium groundwater from Tables I and II are shown in Figure 4.

The modeling results reflect the individual behavior of the uranium concentration in each well. That is, although there are high total specific activities throughout the watershed, you may have the status of two neighboring wells, in which one of them the specific activity is above the permissible concentration limits set by the regulation [10] and another showing concentration below it.

TABLE I. Total uranium specific activity from wells reported in [1]. The absolute uncertainty is expressed in  $\pm\sigma$

Well	$A_{sp}(\text{Bq/L})$	$\sigma_{abs}(\text{Bq/L})$
M 2	0.47	0.08
P 3	0.57	0.07
P 4	1.22	0.05
SN 4	0.66	0.07
SN 6	1.02	0.07
SN 8	0.76	0.06
SF 1	0.61	0.07
F 1	0.98	0.06
C 8	0.98	0.06
C 7	0.57	0.07
V 1	0.90	0.06

TABLE II. Total uranium specific activity from wells from this sampling. The absolute uncertainty is expressed in  $\pm\sigma$

Well	$A_{sp}$ (Bq/L)	$\sigma_{abs}$ (Bq/L)
<b>M2</b>	0.37	0.03
SN 1	0.30	0.02
SN 2	0.47	0.03
SN 3	0.55	0.04
<b>SN 4</b>	0.43	0.03
SN 5	0.50	0.04
SN 7	0.48	0.04
CM 1	0.59	0.05
SV 1	1.60	0.16
SV 2	0.40	0.03
SV 3	0.47	0.01
SV 5	0.28	0.02
VD 1	0.44	0.03
VD 2	0.44	0.03
PC 1	0.69	0.06
PC 2	0.65	0.05
PC 3	0.52	0.04
A 20	0.56	0.04
<b>C 8</b>	0.96	0.05
<b>C 7</b>	0.6	0.05
<b>V 1</b>	0.64	0.05
<b>F 1</b>	0.85	0.09
RV 1	0.33	0.02
NDA 2	0.31	0.02

Notice: Duplicated wells from the 2003 sampling are shown in bold.

3.2. Aquifer host rocks radioactivity results

Table III presents the results of the activity of  $^{238}\text{U}$  divided by the total mass of sediment ( $^{238}\text{U}/m_{tot}$ ) for each sample, depending on its depth. Samples of drilling fragments and powder from Torreoncillos y RS 6 water wells are given.

Here were used only the results of Standard Striping program in which the fitting had a  $\chi^2 < 2.3$ . Figure 5 shows the  $^{238}\text{U}/m_{tot}$  ratios in terms of the depth of each sample for Torreoncillos well. Figure 5 shows also a trend curve, obtained by spline, of  $^{238}\text{U}/m_{tot}$ . This trend line represents the variability of the content of  $^{238}\text{U}/m_{tot}$  in strata (see below).

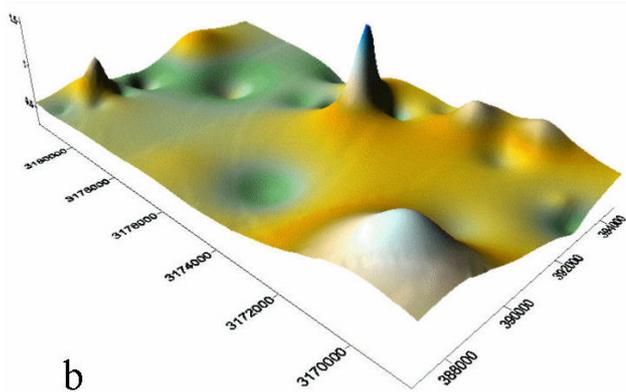
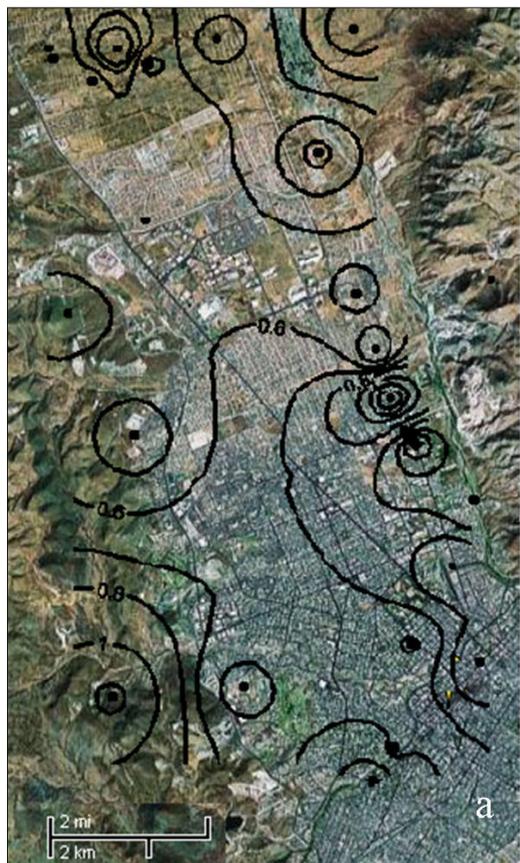


FIGURE 4. (a) Results of water total uranium activity concentrations in the form of isoactivity lines; (b) surface representing the total uranium specific activity in groundwater in Chihuahua-Sacramento subbasin. The surface ordinates and isoactivity lines corresponds each other. Coordinates in (a) are the same as in Figure 2. Coordinates in (b) are in UTM system.

TABLE III.  $^{238}\text{U}$  activity of the sediment fine fraction divided by the total mass of the sample in wells Torreoncillos and RS 6 at different depths. SCL means sandy-clay lacustrine, SCS-sandy-clay soil, and RC-rhyolitic conglomerate. The absolute uncertainty is expressed in  $\pm\sigma$ .

Torreoncillos		
Depth (m)	Texture	$^{238}\text{U}$ (Bq/kg)
50	SCL	181±3
60	SCL	456±8
70	RC	439±10
110	RC	158±2
120	RC	523±11
130	SCL	608±14
140	RC	602±12
150	RC	374±6
170	RC	202±3
180	SCL	418±7
190	SCS	481±10
210	RC	249±3
220	RC	439±9
230	RC	270±4
250	RC	237±4
RS 6		
4	RC	510±13
8	RC	413±7
12	RC	755±19
34	RC	203±3
52	RC	322±6
96	RC	268±4
100	RC	531±11
124	RC	497±10
150	RC	476±9
154	SCS	533±11
162	SCL	362±7
174	SCL	463±9
182	SCL	460±9
198	SCS	418±8
202	SCS	512±9
204	SCS	391±7
206	SCS	612±13
222	SCS	479±8
232	RC	522±11
234	SCS	399±8
236	SCS	492±9
238	SCS	461±9
240	RC	371±7
250	RC	406±7
252	RC	379±6
258	RC	447±9
274	RC	463±8

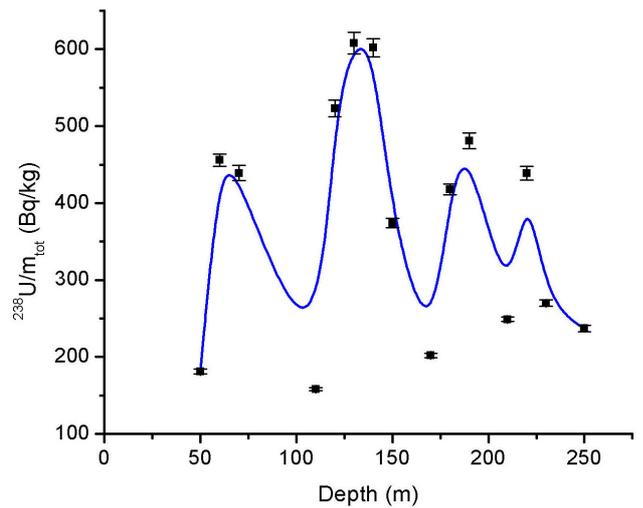


FIGURE 5.  $^{238}\text{U}/m_{tot}$  activity of samples from drilling of well Torreoncillos. Values are shown with uncertainty bars ( $\pm\sigma$ ) and the trend curve is shown as spline (blue), depending on the depth of the sample.

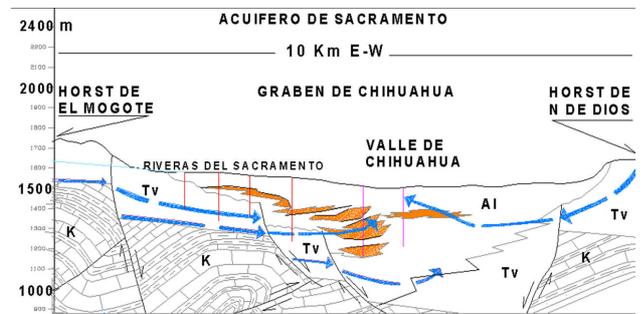


FIGURE 6. Conceptual model for precipitation and concentration of uranium in the subsoil of the Chihuahua-Sacramento Valley: Deep wells in red lines, sandy clay horizons with carbonaceous material in orange, groundwater flow in blue. Ai - Alluvial, Tv - Tertiary volcanic rhyolite, K - Cretaceous.

### 4. Discussion

In the two wells studied by GS were found associations between the  $^{238}\text{U}/m_{tot}$  values and the presence of clays. Although uranium concentrations were not high enough to identify mineralogical varieties through XRD or optical microscopy, radiometry indicates relatively high uranium content in some of the sandy-clay strata, as shown in Table 4 and Figure 5. The abnormal sections examined showed variable contents of carbonaceous material that could be derived from floodplain sediments and paleo-soil. It is inferred that clays play a reducing role by its content of carbonaceous material or  $\text{H}_2\text{S}$ , so that uranium can be precipitated as  $\text{U}^{+4}$  ion [26]. Furthermore, Eisenbud and Gessel have pointed out that an essential characteristic of the platelike particles of secondary aluminum silicates that comprise clay is the abundance of negative surface charges. The resultant ability of the clay particles to attract ions, especially positive ions, to their sur-

faces is one of the most important properties of soils [27]. This way, uranyl cation is adsorbed on clay particles, also.

As a result, the uranium is immobilized in the clay strata. The analysis of the variability of the total uranium activity in groundwater (section 3.1) and the activity of uranium in sediment strata extracted from wells leads, together with the results published in [12, 28], to propose a conceptual model outlined in Figure 6. Water wells at northern Chihuahua-Sacramento Valley, and presumably throughout the valley, occasionally cross lenses of clay strata having deposited uranium. The lenses are the result of formation in the last million years of soil and lake bottom, where uranium transported in solution and suspended matter [28] has been reduced and has remained immobilized. Later a more oxidizing groundwater with bicarbonates redissolves and desorbs uranium, and this water is extracted by the pumping in wells. Overexploitation of the aquifer in the valley southerly direction favors the flow of water from recharge areas with low uranium content, which contributes to the dissolution and desorption of uranium at the clay lenses. Geological conditions for Figure 1 and 6 are extracted from [29]. The characteristics of the process that we suggest is better described below.

The uranyl ion can be in solution while it is at an oxidizing environment. When the water flow transits from an oxidizing environment to a reducing one the uranyl U (VI) is deposited as U (IV) temporarily until new oxidizing solutions transform uranium from the reducing zone. According to [30], primary uranium (as it would be coming from original site in San Marcos area) can be mobilized by oxidation to hexavalent state by the reaction:



Subsequently, the hexavalent uranyl, carried by the flow of surface water and groundwater, enters a reducing zone and is precipitated again as tetravalent. Langmuir [30] has pointed out that reduction of mobile  $UO_2^{2+}$  species to highly insoluble  $UO_2$  must reflect the concurrent oxidation of proportionate amounts of more abundant species of, for example, iron, sulfur and/or carbon.

Several authors [31-33] have described the fractionation of  $^{234}U$  and  $^{238}U$  isotopes in solution at natural environments. They describe the contribution from each isotope to solution through rock dissolution, radioactive decay of parent and daughter, adsorption and desorption of both two isotopes. These three studies describe the redox fronts which are nearly stationary, produced by the arrival of solutions or uranyl complex to reducing areas. Due to the phenomenon known as "recoil",  $^{234}U$  atom is more soluble (and form uranyl ions) [34-38]. Recoil ejection [39, 40] is a combination of the atom extraction resulting from alpha decay (in our case, the  $^{238}U$  product, *i.e.*  $^{234}Th$ ) off its site in the molecule or in crystal lattice in which was the parent atom, and the possible oxidation by Szilard-Chalmers effect in the  $^{234}Th$ - $^{234}Pa$  decay up to form  $^{234}U$ . As a result, in the vicinity of the redox front in groundwater is a  $^{234}U/^{238}U$  activity ratio (AR) significantly

greater than 1 [31-33]. In the water of three of the wells reported in this study have been reported AR between 5.7 and 6.25 [12].

## 5. Current Works

- Mobility of uranium in surface water, vadose zone and groundwater in the San Marcos basin, Chihuahua, Mexico: The effect of water quality parameters in the transport of uranium in the three types of water in the area, from the reservoir Victorino [11] to the ranch area about 20 km away to the East are being studied.
- Study of the crystallography and the solubility of carnotite and tyuyamunite. These species are abundant in arid fields, such as San Marcos and Peña Blanca. The crystal structure of both compounds on the basis of single crystal diffraction is not reported, due to the difficulty of obtaining them. We study the route of synthesis [41-44] and crystal growth by hydrothermal method and fusion [45-48].
- Study of contamination in fish at San Marcos Dam and other tributaries of the Conchos River, Chihuahua. On the basis of previous work in which high uranium activity has been reported in fish at San Marcos [12] a study to find the contents of polonium and other trace elements is performed in this and other dams in the Conchos River, which is the main river of the Chihuahua state.
- Dating of sediments from the bottom of the San Marcos dam through the activities of  $^{210}Pb$ - $^{210}Po$  [49-51] and relation with trace element contaminants. It is investigated the correlation between the contents of uranium and other contaminants in the San Marcos area, in bottom sediments of the dam which could be up to 50 years old.

## 6. Conclusions

From the total activity concentrations of uranium in water extracted from wells in the Chihuahua-Sacramento Valley is observed that individual values do not correspond to the age or to the well depth, but show an apparently arbitrary pattern. The study of the specific activity of  $^{238}U$  contained in the fine fraction of drilling fragments of wells in the area provides a reasonable correlation between the high content of carbonaceous clays and  $^{238}U$  content. Hydrogeochemical conceptual model explains the dispersion of uranium and water radioactivity in the valley. The hydrogeological origin of radioactivity in Chihuahua includes uranium deposits at San Marcos and possibly other not detected sites in the sierras surrounding Chihuahua valley. Water and suspended material carry uranium to reducing areas, lenses formed by sandy clay

mixed with carbonaceous material, paleo-soils and paleo-flooding sediments. Uranium precipitates in these bodies can be redissolved by oxidizing water flow and then be extracted at drinking water wells. Areas of local enhanced uranium contents could be identified through exploratory boreholes to avoid drilling in areas with radiometric anomalies.

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