Characterization of source rocks and groundwater radioactivity at the Chihuahua valley

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As part of a scientific research project about alpha radioactivity in groundwater for human consumption at the Chihuahua City, the characterization of rock sources of radioactivity around de Chihuahua valley was developed. The radioactivity of groundwater and sediments was determined, too. The radioactivity of uranium- and thorium- series isotopes contained in rocks was obtained by high resolution gamma-ray spectroscopy. Some representative values are 50 Bq/kg for the mean value of Bi-214 activity, and 121.5 Bq/kg for the highest value at West of the city. The activity of sediments, extracted during wells perforation, was determined using a NaI(Tl) detector. A non-reported before uranium ore was localized at the San Marcos range formation. Its outcrops are inside the Chihuahua-Sacramento valley basin and its activity characterization was performed. Unusually high specific uranium activities, determined by alpha spectrometry, were obtained in water, plants, sediments and fish extracted at locations close to outcrops of uranium minerals. The activity of water of the San Marcos dam reached 7.7 Bq/L. The activity of fish, trapped at San Marcos dam, is 0.99 Bq/kg. Conclusions about the contamination of groundwater at North of Chihuahua City were obtained.

Keywords: Radioactivity; groundwater; uranium-series disequilibrium; gamma-ray spectrometry; alpha-ray spectrometry.

Como parte de un proyecto científico sobre actividad alfa en agua subterránea de consumo humano en la ciudad de Chihuahua, se ha desarrollado la caracterización de las rocas fuentes de la radiactividad en el valle de Chihuahua. Se determinó también la actividad de muestras de agua subterránea y de sedimentos. La actividad de los isótopos de las series radiactivas en las rocas se obtuvo usando espectroscopía gamma de alta resolución. Algunos valores representativos son 50 Bq/kg para el valor medio de la actividad del Bi-214 y 121.5 Bq/kg para el valor más alto al oeste de la ciudad. La actividad de los sedimentos extraídos durante la perforación de pozos para agua potable, se determinó usando un detector de NaI(Tl). En la formación montañosa de San Marcos se localizó un depósito de uranio no publicado. Sus afloramientos se encuentran dentro de la cuenca del valle Chihuahua-Sacramento y se realizó la caracterización de su radiactividad. Se determinaron actividades por espectrometría alfa de agua, plantas, sedimentos y peces extraídos en emplazamientos cercanos a los afloramientos de minerales de uranio. La actividad del agua en la presa de san Marcos llegó a 7.7 Bq/L. La actividad de pescados de la misma presa alcanza 0.99 Bq/kg. Se obtuvieron conclusiones sobre la contaminación del agua subterránea al norte de la ciudad de Chihuahua.

Descriptores: Radiactividad; agua subterránea; desequilibrio U-234/U-238; espectrometría gamma; espectrometría alfa.

PACS: 89.60-k; 91.65.Rg; 92.40.kc; 92.40.Gc; 92.40.Q

1. Introduction

The geologic substrate seem to justify the high contents of the radioactive isotopes U-234, U-235, U-238 and Ra-226 preliminarily obtained in other works [1,2] in samples of groundwater, as well as in the soil, and randomly in the air, at the cities of Aldama and Chihuahua, in Northern Mexico. The reported specific activities of uranium in groundwater go from 0.41 to 1.4 Bq/l, whereas the interval of the radium runs from 0.28 up to 2.66 Bq/l. These reported specific activities are high if compared with the Mexican norms for use and human consumption of water that fixes for alpha emitters the limit of 0.56 Bq/l. In other Mexican locations the reported activities for groundwater are much less. Among those, the greater value reported by Olguin et al. [3] in Villa de los Reyes is 0.72 μg/l (equivalent to 0.0018 Bq/L of natural uranium). An investigation in the Valley of Toluca [4] reported that groundwater uranium concentration values, from 10 wells, are below 0.2 μg/l.

Chihuahua City mostly rest on the Chihuahua-Sacramento Valley. The geological investigation of the valley suggests that at least two zones, one at South and the other at Northwest, could contribute with uranium minerals to the al-

luvial deposits. The water drained on the outcrops recharges the aquifers and, in principle, uranium minerals could also be in the basement of the valley.

The interest of studying U-series disequilibria around Chihuahua city has been reflected in other publications. Wong *et al.*[5] have reported on U-series disequilibria among other techniques applied to radionuclide migration associated with a uranium-mineralized breccia pipe hosted in Peña Blanca . The Peña Blanca ore, at the NE of the city, has been exploited since the 80's, and now serves as repository of the old uranium mill parts. Its detailed characterization constitute a big research project as a natural analogue of the proposed US federal waste repository site at Yucca Mountain.

This paper presents U-series isotope activities obtained by high resolution gamma-ray spectroscopy in rhyolitic tuffs and rocks around Chihuahua City. The uranium, thorium and potassium activity of sediments, extracted as fragments during perforation of two wells, was determined using a NaI(Tl) detector. A non-reported before uranium ore was localized at Majalca-San Marcos range formation, at NW. Its outcrops are inside the basin of the Chihuahua valley and its uranium activity characterization was performed in this paper. Uranium isotopes activity of water, plants, sediments and fish sampled near to the San Marcos outcrop was determined by alpha spectrometry, and the activity ratios (AR) of U-234/U-238 were obtained by alpha spectrometry, too.

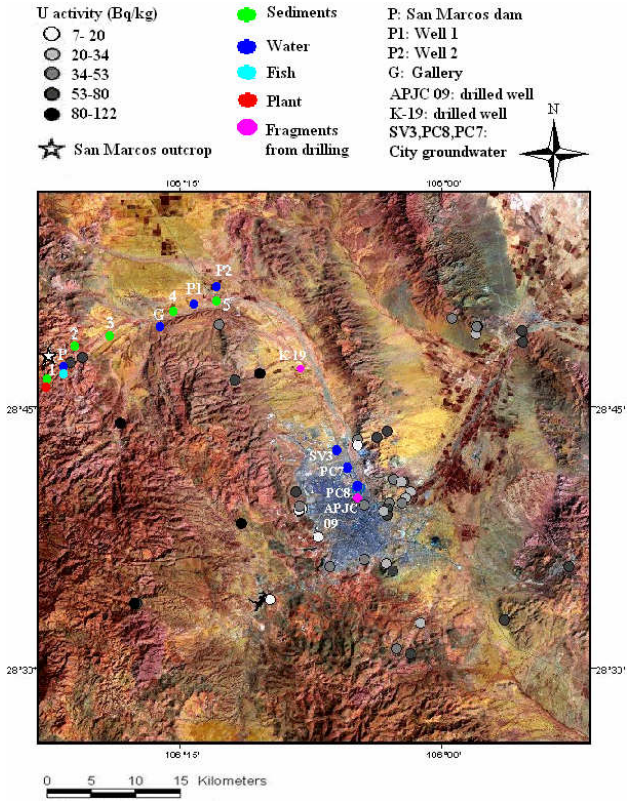

FIGURE 1. Satellite image with the location of water, sediments, fish, plants, and rocks sampling sites in the Chihuahua-Sacramento and the Tabalaopa-Aldama valleys. Activity concentrations of Bi-214 (U-234) in rocks extracted around Chihuahua City are presented in gray scale. Values are presented in Bq/kg of rock.

U-series disequilibria results suggest relationships among different agents of isotope transport and reception, and partially explain the origin of groundwater radioactivity previously reported in Ref. 1.

2. Materials and methods

Figure 1 presents the location of sampling of all specimens analyzed in this paper. More than 50 rock samples were extracted from ranges around the Chihuahua-Sacramento and the Tabalaopa-Aldama valleys and hypothetically forming alluvial sediments. Most of the rocks forming the ranges are rhyolites, rhyolitic tuffs and carbonates from the Tertiary age [6]. The rocks were chosen on the base of the geological information of the zone and by means of spectral methods applied to satellite images. These samples were studied by high resolution gamma-ray spectroscopy.

Alluvial sediments forming the Chihuahua-Sacramento Valley, obtained as fragments from drilling of two new wells, were selected from the South and the North of the valley for the NaI(Tl) gamma-spectroscopy study.

For the alpha spectrometry study and the AR determination the following samples were selected (Fig. 1). Most of the alpha spectrometry was developed at the University of Seville, Spain:

- One rock sample of the outcrop of the San Marcos formation, near to the homonymous dam.
- Water from the dam, from two wells (30 m in depth, approximately) and from a so-called gallery, separated by approximately one kilometer one from the other.
- Five sediment samples, the first near the outcrop and the rest, following the San Marcos affluent of the Sacramento River.
- Fifteen samples of fish from the dam.
- Plants growing close to the outcrop, divided afterward in root, stem and leaves.
- For reference of the water for human consumption, three sites for groundwater samples of wells (100 m in depth, approximately), from the center of the Chihuahua City, were selected and analyzed.

Gamma-ray spectroscopy was performed at CIMAV. The high resolution gamma-ray detector used in this investigation is a cylindrical, coaxial type HPGe. Details of measurements are described elsewhere [7].

For measurements of sediments by gamma spectrometry the selected sample geometry was a cylinder, practically in contact with a BICRON NaI(Tl) detector. All samples, of masses approximately equal to 230 g, were grounded, sealed

and allowed to stand for 4 weeks to establish the secular equilibrium. It was experimentally confirmed that gamma rays absorption coefficients of rocks, sediments and soil depend only on energy and not on material matrix. Efficiency calculation for different heights in the container was performed, using a MonteCarlo code EGS4. Four standard spectra of background, Th-232 series, U-238 series and K-40 were obtained in the selected geometry, using the IAEA Standard Reference Materials RGK-1, RGTh-1 and RGU-1. Sample spectra were obtained similarly. A computer code in Visual BASIC was done, which perform activity calculation with error, by the relative method.

Preparation after sampling of specimens subjected to alpha spectrometry was performed at CIMAV, Chihuahua. All water specimens were acidified using HNO_3 immediately after sampling. Sample volumes were equal to 6 L. Alpha spectrometry of groundwater samples PC7, PC8 and SV3 (Fig. 1) from the center of the city was performed at CIEMAT, Madrid. Aliquots of 100-200 mL of the samples were radiochemically purified by anion exchange and solvent extraction with the addition of ^{236}U as yield tracer [8]. The U fractions were electrodeposited onto stainless-steel discs using the method described by Hallstadius [9]. Sources were measured using CANBERRA silicon-implanted detectors of 450 mm^2 active area in TENNELEC TC 257 alpha spectrometers. The good resolution of the spectra allowed the measurement of the area under the peaks by simple integration. The small contribution of ^{235}U under the peaks of ^{236}U was corrected.

Digestion and preparation of samples, as well as alpha spectrometry of the rest of specimens were performed at the University of Seville. Before being put under the radioanalytical analysis procedure, the aliquots of water samples extracted from San Marcos site were spiked with known amount of U-232. Plant, fish and sediment samples were burned. Their ashes were also spiked with U-232. Total sample dissolution was performed by acid digestion. In the case of sediment samples, digestion was made using aqua regia, and for the plant and fish samples, total dissolution was made using H_2O_2 and 8M HNO_3. Radioanalytical procedure performed on all liquid samples was the following: After a coprecipitation with $Fe(OH)_3$, uranium was isolated from the sample by solvent extraction with TBP [10]. Then, uranium was electroplated for alpha-spectrometric analyses [11]. An alpha-spectrometry chain Alpha Analyst (CANBERRA) was used for alpha activities measurements. Radiochemical yield was determined with ^{232}U information.

3. Results and discussion

3.1. Gamma-ray results

In Table I, the specific activities of uranium and thorium series, and the activity of K-40 of rock samples with highest values of U-series activities are shown. Figure 1 presents, in gray scale, the specific activity of uranium series of all rock samples.

From Table I and Fig. 1 it is observed that most of highly radioactive rocks are located at the North and West of the Chihuahua-Sacramento Valley. Over there has been observed also most of the rhyolitic rocks with abundant silica.

TABLE I. Specific activities, in Bq/kg, of K-40, U- and Th-series radioisotopes and activity ratios of rock samples with the highest values of U-series activities.

Sample	Th-234	Pb-214	Bi-214	$\frac{\text{U-234}}{\text{U-238}}$	K-40	Ac-228	Pb-212	$\frac{\text{Th-232}}{\text{U-234}}$
LHE14	107.6	121.9	117.8	1.10	1354	53.5	58.6	0.45
LHW18	149.0	126.2	121.5	0.82	1638	145.4	147.3	1.20
SAN21	145.3	105.9	101.2	0.70	1810	157.9	157.3	1.56
SME26	160.0	99.0	95.9	0.60	1332	161.5	162.0	1.68
SME27	103.4	109.3	105.7	1.02	1375	72.1	68.5	0.68
SME28	111.6	75.2	72.5	0.65	1939	142.7	143.2	1.97
SME29	105.0	83.3	79.6	0.76	1819	114.7	114.9	1.44
JME30	144.7	104.6	100.5	0.69	1338	155.0	155.6	1.54
SES40	111.5	76.2	72.6	0.65	1030	95.8	99.0	1.32

TABLE II. Geometric mean values of specific activities for natural uranium, thorium and K-40 of fragments extracted while drilling at north and south of Chihuahua-Sacramento Valley.

	Uranium (Bq/kg)	Thorium (Bq/kg)	Potasium-40 (Bq/kg)
K_19	54.78	272.41	1392.49
APJC_09	63.20	111.81	1555.83

Most specific activities of U- and Th-series presented in Table 1 are less than or of the same order as those atributed to rhyolites, which have contents of uranium from 1 to 18 ppm (approximately from 12 to 230 Bq/kg) [12]. Radioactivity of rhyolites is explained as a result of the slow diffusion of large U- and Th- series cations in melting and solidifying magma. Thus, in spite of the incompatible character of those cations, probably the processes don't occur in equilibrium and large cations remains in the solid part [13].

It may be observed from Table I, that the values of the AR U-234/U-238 are less than 1. It implies that in general parent and daughter are not in equilibrium. It is considered, nevertheless, that these results may have a bias due to the possible contribution of the Th-232 family x-rays in the line of 92.6 keV [7], which would increase its intensity. Major part of analyzed rocks are Tertiary rhyolites and are homogenous. The content of radioactive isotopes is attributed to the rock matrix, and due to their ages, samples must be in radioactive equilibrium.

In Table II, the geometric mean values of the specific activities of natural uranium, thorium and K-40 of the sediment fragments extracted while drilling of two wells are presented. Each sample represents an approximated interval of 4 meters. For well APJC-9, at the south (Fig. 1), 51 samples were analyzed, representative of the depth intervals from 0 to 150 m and 400 to 500 m, in discontinuous way. For well K-19, to the north, 31 samples were analyzed, representative of the depth interval from 88 to 206 m, almost continuosly studied. With the NaI(Tl) detector and the method exposed above a good distinction of uranium concentration was obtained at the different profile zones. All random uncertainties of specific activities were less than or equal to 7%.

The behavior of the radioactivity profiles of the wells APJC-09 and K-19 may be explained from the geologic history of the valley. Thousands years ago the Chihuahua-Sacramento basin was closed to the Southeastern side (Coronel-Nombre de Dios). Over there is the lowest level in the basin, the Sacramento and Chuviscar rivers flow and converge there (Fig. 1). In one hand, the samples extracted from well APJC-09, to the south, present greater uranium content. The K-19 well, at the north, shows greater average in thorium concentration. On the other hand, the rock samples from San Marcos and from Cerro Jose Maria (Fig. 1, SME and JME in Table I) are the most radioactive in Th and U. The correlation of the specific activities determined in samples from well APJC-09, as much for uranium as for potassium, shows the presence of rocks from formation of the entire valley. It is suggested that when the basin was closed, the zone of well APJC-09 (South) captured eroded rock sediments and waters with great uranium content from the north. Meanwhile, by the Northern zone remained the rocks more enriched in thorium, which is not soluble, that are represented in the K-19 well.

TABLE III. Specific activities of U-238, U-234 isotopes, and activity ratios between them, for groundwater samples extracted from Chihuahua City wells. Absolute errors are shown in parenthesis.

City groundwater well	U-238 (Bq/L)	U-234 (Bq/L)	U-234 / U-238
PC7	0.086 (0.004)	0.534 (0.026)	6.25
PC8	0.138 (0.004)	0.792 (0.026)	5.73
SV3	0.065 (0.002)	0.405 (0.012)	6.24

TABLE IV. Specific activities of U-238, U-234 isotopes, and activity ratios between them, for water samples extracted from sites, close to San Marcos outcrop. Absolute errors are shown in parenthesis.

Water	U-238 (Bq/L)	U-234 (Bq/L)	U-234 / U-238
Dam	7.70 (0.36)	34.78 (0.94)	4.52
Gallery	1.97 (0.23)	4.10 (0.15)	2.08
Well 1	6.75 (0.45)	27.03 (1.04)	4.00
Well 2	6.07 (0.40)	21.20 (0.84)	3.49

TABLE V. Uranium isotopes specific activities and activity ratios values of sediments of San Marcos affluent. Absolute errors are shown in parenthesis.

Sediment sample	U-238 (Bq/kg)	U-234 (Bq/Kg)	U-234 / U-238
M1	23.04 (0.94)	34.13 (1.23)	1.48
M2	19.64 (0.76)	15.94 (0.67)	0.81
M3	16.35 (0.68)	19.89 (0.77)	1.22
M4	9.88 (0.36)	10.39 (0.37)	1.05
M5	16.58 (0.52)	15.64 (0.50)	0.94

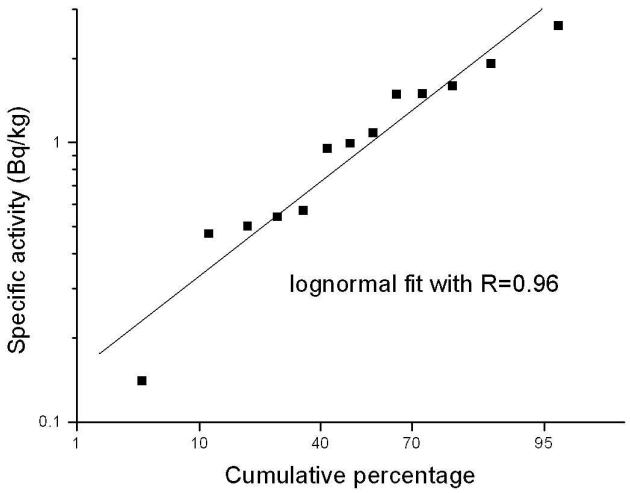

FIGURE 2. Probability graph of U-238 activity concentrations for fish sampled at the San Marcos dam. Straight line represents the lineal fit of the experimental values.

TABLE VI. Uranium isotopes specific activities and activity ratios values of plant sections and the uranium mineral from the San Marcos outcrop. Absolute errors are shown in parenthesis.

Plant	U-238 (Bq/kg)	U-234 (Bq/Kg)	U-234/U-238
Root	1.69 (0.07)	1.89 (0.07)	1.12
Leaf	5.28 (0.12)	5.83 (0.13)	1.10
Stem	1.10 (0.03)	1.08 (0.03)	0.98
S M outcrop	1538500 (29200)	1529300 (29100)	0.99

TABLE VII. Uranium isotopes specific activities and activity ratios values of fish samples, captured in San Marcos dam. At bottom of the table are presented the statistical parameters of the distributions for U-238 and U-234, respectively. Absolute errors are shown in parenthesis.

Fish	U-238 (Bq/kg)	U-234 (Bq/Kg)	$\frac{\text{U-234}}{\text{U-238}}$
P1	0.95 (0.21)	1.99 (0.32)	2.10
P2	1.59 (0.19)	2.60 (0.24)	1.63
P3	1.91 (0.33)	1.70 (0.30)	0.89
P4	1.48 (0.11)	2.71 (0.15)	1.82
P5	1.08 (0.09)	1.78 (0.12)	1.66
P6	2.61 (0.16)	4.54 (0.22)	1.74
P8	0.50 (0.09)	1.43 (0.14)	2.88
P9	0.57 (0.08)	1.17 (0.11)	2.06
P10	0.47 (0.07)	1.63 (0.14)	3.46
P12	0.14 (0.06)	0.63 (0.11)	4.48
P13	1.49 (0.11)	1.99 (0.12)	1.34
P15	0.99 (0.12)	2.10 (0.18)	2.12
P16	0.54 (0.15)	2.00 (0.27)	3.69
Mean value	1.1	2.02	
Geom mean	0.88	1.84	
Mode	0.99	1.99	

3.2. Alpha spectrometry results

Table III, shows specific activities of U-238 and U-234 isotopes, as well as AR U-234/U-238, for groundwater samples extracted from Chihuahua City wells. Figure 2 shows alpha spectrum of one of the samples. It is noticeable the strong disequilibrium observed between uranium-series isotopes.

Table IV, reflects the results of alpha spectrometry for water sampled near to the San Marcos outcrop.

The values of specific activity of uranium isotopes obtained in the San Marco's zone are high. Resulting AR values show an interval from 2 to 4. Chabaux *et al.* [14] and Porcelli and Swarzenski [15] discuss the causes of disequilibrium of both isotopes in surface and groundwater, respectively. Several, related to each other, mechanisms are exposed: in rocks, U-238 preferably exists in its oxidation state +4 that is practically insoluble in water. Its disintegration produces Th-234, which undergoes two successive beta decays. The recoil of all these disintegrations produces the electron stripping of U-234 ion that oxidizes to hexavalent state. The U+6 is soluble in water as uranyl ion. Also it has been suggested that since U-234 is resident in damaged lattice locations, it is more vulnerable to oxidation by fluids. Alternatively, it has been suggested that as U is released by weathering, tetravalent U-238 is preferentially precipitated or adsorbed, while hexavalent U-234, oxidized during the recoil process, more readily remains in solution [15]. This will increase the AR of the groundwater if isotopic equilibration does not then occur between adsorbed and dissolved U. These arguments are valid for both dam and groundwater sampled in sites near to the San Marcos outcrop. A linear correlation could be found between the contents of U-238, U-234 and AR values obtained in the samples versus distance to the outcrop, but the gallery sample presents figures twice lower than the others. These lower values respond to the characteristics of the superficial water in constant movement. Osmond and Ivanovich [16] explains this behavior by the mixture of runoff water and diverse groundwater sources. In this case, the superficial filtered water is in contact with groundwater of the uraniferous environment of the San Marcos area.

Table V shows uranium isotopes specific activities and AR values obtained from sediments sampled from the San Marcos affluent of the Sacramento River. Specific activities become lower while the sampling site is farther from the San Marcos outcrop. The obtained values are relatively low, and could be explained by adsorption of uranium during the water runoff. The U-234/U-238 AR is close to one, answering to the same reasons given in the preceding paragraph [15].

Table VI, presents uranium isotopes specific activities and AR values obtained for the yellow dust scrapped from veins present in a rhyolitic rock sampled from the outcrop of the San Marcos site. Also are presented the results obtained for the plants growing near to the outcrop, divided afterward in root, stem and leaves.

Yellow dust from the San Marcos outcrop was identified by XRD [17]. The mineral composition was mainly uranophane, uraninite, quartz and some hematite. This composition justifies the high uranium activity observed. The equilibrium value of the AR implies that the mineral deposits has an age equal to or grater than one million years [13].

The high values of specific activity in leaves are not surprising. UNSCEAR 2000 [18] reports greater activity con-

centration of U-238 in leafy vegetables that in fruits and root vegetables. Migration and accumulation of radionuclides in the soil-plant system is complex. The radionuclides concentration in soil doesn't determine alone their concentration in plants [19]. Factors such as soil characteristics, climatic conditions, type of plants, part of the plant concerned, physicochemical form of the radionuclides and the interfering element can all influence the transfer factor values. In one hand, Chen *et al.* [20] have obtained root/shoot ratios for different plants for U-238 from 2.8 to 16.2. On the other hand, Ould-Dada *et al.* [21] have found greater radionuclide activities in leaves that in fruit. They explain that the activity in leaves is due to the contribution of several processes. These processes can be interception, absorption, resuspension and translocation from roots to other components of the plant. Therefore, the obtained values demonstrate the possibility of uranium contamination in agriculture but are not categorical to be a reason for public health concern.

In Table VII, the U-238 and U-234 specific activities of clean muscle from fish, captured in San Marcos dam are presented. Activity ratios of U-234/U-238 in samples are about 2 or more. This result was expected, because fish live and drink in the water with AR equal to 4. The obtained values of specific activity were studied statistically. Lognormal distributions for both isotopes were obtained. Of the 15 samples, two activity values were below the detection limit, and they were not used in the statistical calculations. Figure 2 presents the probability graph of U-238 activity concentrations, showing the lognormal character of the distribution. Reported values in UNSCEAR 2000 [18] for the United States have an interval of specific activities for U-238 from 0.013 to 1.9 Bq/kg. Any of the statistical parameters of the lognormal distribution for U-238 obtained in this study are much greater than the reference value of 0.1 Bq/kg reported in UNSCEAR 2000. These facts emphasize the abnormal concentration of uranium in water and fish, and suggest giving that information to the public.

4. Conclusions

The higher natural radioactive isotope concentrations are at the northwest of the Chihuahua-Sacramento Valley, mainly in the mineralized deposits of San Marcos. This zone to a great extent contributes to the high contents of uranium and thorium observed in the alluvial deposit, as well as in superficial and ground waters. The great values of activity ratio U234/U238 in water, not only near the outcrops, but also in the city, indicate that the Chihuahua valley is almost totality a uraniferous zone. The specific activity in water samples is greater than the limit for alpha emitter, allowed by the Mexican official norm of water quality for human consumption. By all these considerations it is concluded that there is a natural contamination by uranium in the different sample types analyzed from the Chihuahua valley.

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