Application of the alpha spectrometry for the study of core sediment extracted in the San Marcos dam in Chihuahua

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The determination of the specific activities of ^{210}Po and isotopic uranium by alpha spectrometry was performed in a sediment core from San Marcos Dam.
The objective of this work was to analyze the vertical distribution of isotopic uranium and ^{210}Po and the behavior of these radionuclides along sediment core collected from the San Marcos Dam. Sample was divided into 11 sections, in which ^{210}Po and Isotopic Uranium were determined using liquid-liquid extraction with tributyl phosphate (TBP). Furthermore, it was made a comparison between the TBP technique and the technique of extraction chromatography using UTEVA resins for uranium.
The results of specific activities for ^{210}Po show a trend to decrease along the core, whereas the uranium isotopic does not show a pointed trend. The results of isotopic ratios between the ^{234}U and ^{238}U show that they are close to secular equilibrium in each of the core sections. The comparison between the two uranium extraction techniques indicates that chemical yield is better using the UTEVA technique than TBP extraction.

Keywords: alpha spectrometry; uranium; polonium; sediment core.

La determinación de las actividades específicas de ^{210}Po y uranio isotópico mediante espectrometría alfa fue realizada en un núcleo de sedimento de la Presa San Marcos. El objetivo de este trabajo fue analizar la distribución vertical de uranio isotópico y de ^{210}Po así como el comportamiento de estos radionúclidos a lo largo del núcleo de sedimento recolectado en la presa San Marcos. La muestra fue dividida en 11 secciones. Para la determinación de ^{210}Po y de uranio isotópico en cada una de las secciones se utilizó la técnica de extracción líquido-líquido con tributil fosfato (TBP). También se realizó una comparación entre la técnica del TBP y la técnica de cromatografía de extracción con resinas UTEVA para el uranio isotópico. Los resultados de las actividades específicas para ^{210}Po muestran una tendencia decreciente a lo largo del núcleo, mientras que para uranio isotópico muestran una tendencia no monótona. Los resultados de las relaciones isotópicas entre el ^{234}U y el ^{238}U muestran estar cerca del equilibrio secular en cada una de las secciones del núcleo. La comparación entre las dos técnicas de extracción de uranio indica que se tiene un mayor rendimiento químico mediante empleando la técnica de UTEVA.

Descriptores: Espectrometría alfa; uranio; polonio; núcleo de sedimento.

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1. Introduction

In the state of Chihuahua there are known about 30 uranium anomalies [1], almost all located in the vicinity of Chihuahua City, the state capital, at the center of the homonym state in Mexico. At north-west of Chihuahua City, Majalca Area, are two well-established major uranium deposits: Victorino and San Marcos, both in the San Marcos Area. There is also the San Marcos dam, namesake of the surrounding mountain. The San Marcos mountain formation is a caldera [2], which includes several outcrops of uranium ore and the dam. Geological investigations suggest that these uranium ores contribute to uranium deposits in sediments at the Chihuahua-Sacramento Valley [3-5].

Previous studies in the area have shown the presence of uranium in superficial waters, fish, and some plants [6-9]. Data about the superficial water contamination shows high uranium specific activities by up to 7.7 Bq/L [10]. This value is above to the reference level of 0.001 Bq/L for drinking water reported in UNSCEAR [11]. The high uranium concentrations in the area are attributed to the lixiviation and/or erosion of the uraniferous deposits located in San Marcos [3, 7].

As foregoing mentioned, there are several studies in different matrices in the study area, but there have been no determinations of radionuclides in a sediment core. The importance of determining uranium in this work is to know its behavior in different sections of the sample, corresponding to different sedimentation depths in the dam. Moreover, the determination of ^{210}Po will allow a future dating work. Different uranium extraction procedures were tested for determining the best parameters for future works.

2. Materials and methods

2.1. Study area

The San Marcos River, which rises in the dam of the same name, is tributary to the Sacramento River in Chihuahua City. The San Marcos River basin covers approximately 175 km^2.

FIGURE 1. San Marcos Dam study area. Sediment core was collected at point B.

Figure 1 shows a satellite image from San Marcos dam; point B shows the place where core was extracted. The dam wall is showed as an angle, which can be seen close to points 5 and B.

2.2. Sediment Core Sample

Figure 2 shows the sediment core sample extracted from San Marcos Dam.

Sampling of sediment core was carried out in July 2007. The core length was 30 cm, and it was divided into 11 sections of 2 – 4 cm width, the sample closest to the surface was designated as MTA1 and MTA 11 is the deepest fraction; each section had a mass about 50 g. In the laboratory, samples were dried at 55 ˚ C during 24 hours as general procedure.

2.3. Determination of radionuclides by alpha spectrometry

For the determination of both^{210}Po and uranium extraction the technique of liquid-liquid extraction with tributyl phosphate (TBP) was used. This method was initially developed by Holm and Fukai [12] and slightly modified by El-Daoushy et al [13]. To obtain the best parameters, the technique of uranium by extraction chromatography using UTEVA resins was tested for comparison.

FIGURE 2. Sediment core sample.

The samples were spiked with ^{209}Po standard solution for the determination of ^{210}Po, and for isotopic uranium determination a solution standard of ^{232}U was used.

Isotopic uranium and ^{210}Po contents of each section of the simple were determined by alpha spectrometry using a spectrometry chain Alpha Analyst (CANBERRA), based on PIPS type detector.

2.4. Extraction of isotopic uranium and ^{210}Po by tributyl phosphate (TBP)

For this procedure, samples were dried at 55^{o}C, sieved through a 2 mm mesh and homogenized. Aliquots of 0.5 g from every core sections were taken. Samples were digested in a hot plate with aqua regia and the dry residue was redissolved using $HNO_3$8M.

Afterwards, the extraction of polonium and uranium was carried out with TBP. The ^{210}Po alpha source is obtained by self-deposition onto copper discs from a 1.5M HCl solution [13], while the final solutions of U, after their isolation, were electrodeposited onto stainless-steel discs by the Hallstadius (1984) [14] method.

2.5. Extraction of isotopic uranium by extraction chromatography with UTEVA resins.

For the extraction of isotopic uranium by UTEVA, samples were calcined at 600^{o}C during 24 hours. After calcination, the samples were sieved to 2 mm mesh and homogenized.

Aliquots of 0.2 g were taken. Samples were digested with HF and $HN0_3$, and then the dry residue was dissolved with 3 M HN03. The sample solution was passed through the UTEVA column. In this condition, U (VI) is retained. Uranium is eluted with HCl 0.01 M, and then it is electrodeposited.

3. Results and discussion.

Table 1 shows the results of specific activities of ^{210}Po and the obtained chemical yield for TBP technique. Figure 3 plot the specific activities of ^{210}Po, and shows that the specific activity of ^{210}Po decreases when the core fraction of sediment is deeper, as was expected.

The result of the specific activity of ^{210}Po in the MTA1, is that corresponding to the fraction of lead in excess.

Figure 4 shows alpha spectra characteristic for polonium and uranium respectively. Note the peak corresponding to polonium extracted from the sample ^{210}Po and the peak of the tracer ^{209}Po (a). The peaks corresponding to uranium isotopes from the sample and ^{232}U peak of the tracer are shown in (b).

TABLE I. Specific activities of ^{210}Po obtained in fractions of the core sample.

CODE	^{210}Po (Bq/kg)	YIELD (%)
MTA 1	167 ± 6	55
MTA 2	200 ± 6	59
MTA 3	183 ± 6	56
MTA 4	162 ± 5	60
MTA 5	148 ± 6	43
MTA 6	133 ± 5	60
MTA 7	134 ± 4	53
MTA 8	107 ± 3	51
MTA 9	89 ± 3	59
MTA 10	89 ± 4	58
MTA 11	84 ± 3	54

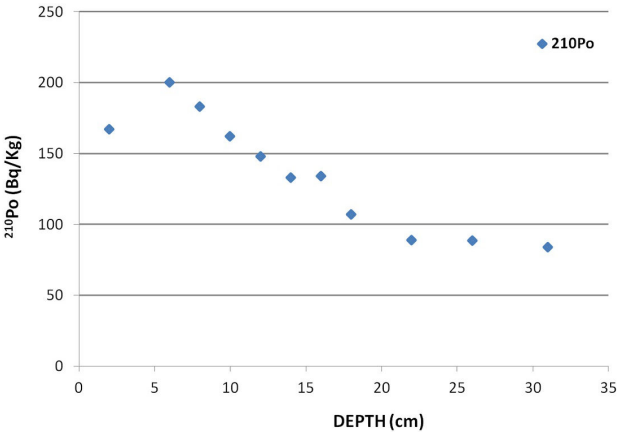

FIGURE 3. ^{2}10Po activity as function of sample depth.

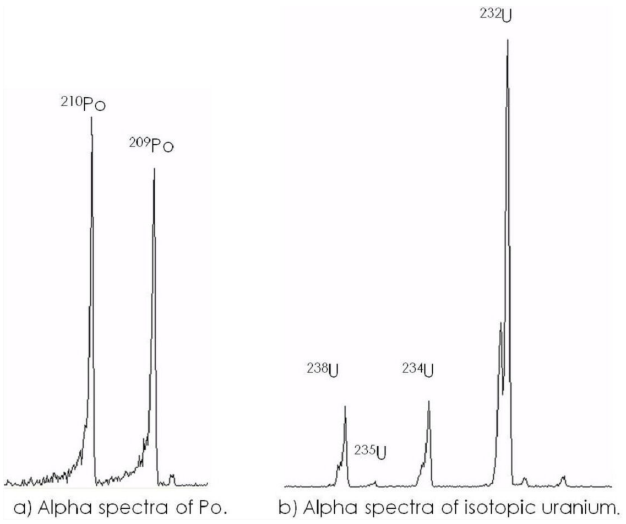

FIGURE 4. Alpha spectrum of Polonium and uranium respectively.

TABLE II. Specific activities of ^{234}U and ^{238}U, and activity ratios in each fraction extracted by UTEVA.

CODE	^{234}U (Bq/kg)	^{238}U (Bq/kg)	(^{234}U/^{238}U)	YIELD (%)
MTA 1	125 ± 9	107 ± 8	1.2	58
MTA 2	126 ± 8	93 ± 6	1.4	71
MTA 3	102 ± 7	79 ± 5	1.3	76
MTA 4	100 ± 7	79 ± 5	1.3	79
MTA 5	119 ± 8	92 ± 6	1.3	80
MTA 6	108 ± 8	95 ± 7	1.1	69
MTA 7	125 ± 9	106 ± 8	1.2	75
MTA 8	119 ± 8	98 ± 7	1.2	76
MTA 9	120 ± 8	96 ± 7	1.2	77
MTA 10	107 ± 7	94 ± 6	1.1	84
MTA 11	92 ± 6	83 ± 6	1.1	81

Table 2 shows the results obtained by UTEVAS technique for isotopic uranium.

Results from Table 2 and Fig 5 show that the ^{234}U and ^{238}U activity ratios are close to secular equilibrium along the sediment core. This behavior probably means that uranium content of sediments comes from suspended particulated matter instead of precipitation from dissolved uranium content in water. The results of the activities for ^{235}U were excluded in this report, due to the high errors found in the measurements.

Table 3 shows the comparison between the two uranium extraction techniques. Results indicate that chemical yields are better using the technique of UTEVA extraction. This technique would be the best for sediment core uranium analysis in future work.

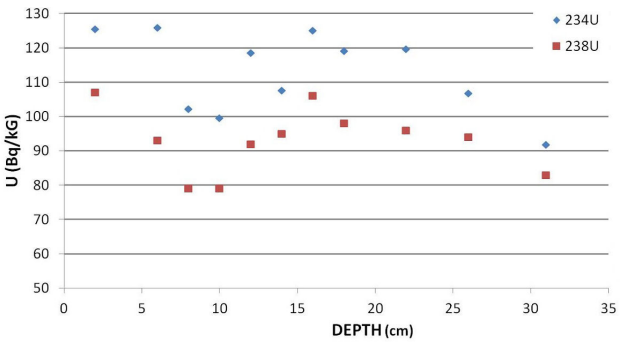

FIGURE 5. Isotopic Uranium activity as function of sample depth.

TABLE III. Comparation between techniques for uranium extraction.

CODE	UTEVA	TBP	UTEVA	TBP	UTEVA	TBP
MTA 1	126	67	107	38	58	55
MTA 4	100	66	79	19	79	59
MTA 5	119	112	92	39	80	55
MTA 7	125	82	106	51	75	52
MTA 9	120	60	96	36	77	48

4. Conclusions

Specific activities of isotopic uranium and ^{210}Po were determined by alpha spectrometry.

In sediment core, the highest concentration of ^{210}Po is in the samples near to the surface, whereas the lowest concentration is in the deeper fraction, as expected.

The behavior of ^{238}U and ^{234}U activities is close to secular equilibrium, as expected in case of sediment formation from solid fragments.

The extraction technique by UTEVA resins gives higher chemical yields for isotopic uranium analysis.

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