# Radon behavior in springs and wells around Cuitzeo lake, Lerma river basin, Mexico

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#### RESUMEN

Se determinó radón en manantiales y pozos en zonas urbanas y agrícolas alrededor del lago de Cuitzeo, en la cuenca del río Lerma, México. Se estudiaron también los elementos mayores y traza. El  $^{222}$ Rn se midió por el método de centelleo líquido, los elementos mayores con análisis químicos convencionales y los elementos traza por espectrometría de masas (ICP-MS). Los valores promedio de las concentraciones de radón oscilaron entre 0.88 y 3.66 Bq L<sup>-1</sup>, indicando un tránsito rápido entre la zona de recarga y el afloramiento en los manantiales. Los valores de los elementos mayores y traza se discuten considerando las características geológicas de los sitios en estudio.

PALABRAS CLAVE: Zona volcánica, agua subterránea, radón, composición química, elementos traza disueltos.

#### ABSTRACT

Radon was determined in springs and wells from urban and agricultural zones around Cuitzeo lake, in the Lerma river basin, Mexico. Major and trace elements were also studied in the water samples. The measurement techniques included the liquid scintillation method for <sup>222</sup>Rn, conventional chemical analysis for major components and ICP-MS for metallic trace elements. The average radon concentration values were relatively low, ranging from 0.88 to 3.66 Bq L<sup>-1</sup> indicating a rapid transit from recharge to the output of the springs. The major and trace elements are discussed considering the geological characteristics of the studied sites.

KEYWORDS: Volcanic zone, groundwater, radon, chemical composition, dissolved trace elements.

# INTRODUCTION

The geological characteristics of aquifers are essential parameters generating differences between the concentration levels of physical parameters, chemical species and radionuclides in the water samples (Segovia *et al.*, 1999). Radon gas is considered as one of the natural radioactive elements with greater mobility in the terrestrial crust (Corbett and Burnett, 1997). Conventional chemical analysis of water does not put the emphasis on the study of water dissolved gases; however, their determination is important, due to their capacity to react. In the case of radon, being a noble gas with low interaction capacity, its presence can provide information concerning the radioactive elements present on the rock, the water origin and flow mechanisms; aquifers can be classified according to their <sup>222</sup>Rn concentration (Hamada, 2000). Groundwater may contain high amounts of <sup>222</sup>Rn generated as a product of the <sup>238</sup>U natural radioactive series decay. Once the groundwater flows to rivers or lakes, the radon content significantly decreases due to degassing and limited regeneration since little amounts only of radium are generally dissolved in surface water (Annanmaeki *et al.*, 2000; Kitto and Kuhland, 1995; Paulus *et al.*, 1998).

The assessment of major and trace elements in groundwater is important to reconcile the exploitation of natural resources with the protection of the environment. Pollution of surface waters can percolate to the substrata and contaminate aquifers. Measurement of the mentioned parameters together with microbiological organisms can provide information concerning a given zone's state of pollution. Cuitzeo lake is located in the northern part of Michoacan State in Mexico. The region belongs to the middle part of the Mexican Neo Volcanic belt. The basin is part of the upper Lerma river, whose watershed is one of the most polluted in Mexico (De Cserna and Álvarez, 1995).

With the aim to evaluate the transport of contaminants to the aquifers in the zone, radon, major and trace elements together with biological parameters were determined from several wells and one spring whose waters are used for drinking water supply of the nearby towns around Cuitzeo lake.

## **EXPERIMENTAL**

## The sites

Recent volcanism has occurred at Michoacan State, the youngest volcano, Paricutin, was born in 1943. The main regional geological formations are from Tertiary and Quaternary periods. Michoacan hydrology is composed by the upper Lerma river, the central lakes zone and the Balsas river. The Cuitzeo basin, in the northern part of the State, having 3977 km<sup>2</sup> is one of the largest lakes of the zone. A fork of the Lerma river flows to the Yuriria lake and reaches Cuitzeo in its northern part. The main landform of the sampling zone is formed by the Cuitzeo depression. The weather is moderate with summer rains (May-October) giving an average annual precipitation of 906 mm and a temperature range from 10 to 28 °C.

### Sampling

The sampling sites are located around the Cuitzeo lake, between 101° 10' and 101° 20' W and 19° 58' 00'' and 19° 58' 23'' N at an average altitude of 1850 m. The samples were obtained from Jeruco, Cuitzeo 2, Cuitzeo 3, Copandaro, Panteón and El Salitre boreholes ranging from 60 to 120 m depth, continuously pumped to provide the drinking water supply of the nearby towns. In Los Baños, spring water was also monitored (Figure 1).

Three sampling campaigns of water for radon determination were performed from March to August, 2001. The samples were taken in carefully cleaned HDPE (high density polietylene) containers tightly closed to avoid degassing. Aliquots of 125 mL sample and 10 mL pure toluene were immediately transferred to a separator funnel and vigorously shaken for 5 minutes for radon separation. When the mixture resettled in two phases, the organic phase was transferred into a counting vial to which 10 mL of INSTAGEL scintillation solution was added (Olguin *et al.*, 1993).

One litre water samples were taken in plastic polyethylene bottles for major chemical components such

as Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, total alkalinity and SiO<sub>2</sub>. A 500 mL sample having 3 mL HNO<sub>3</sub> was used for the analysis of Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>.

The sampling for trace elements dissolved in the water was performed in 60 mL HDPE bottles previously washed and decontaminated with  $HNO_3$ : one day before the sampling the bottles and covers were rinsed three times and filled with deionized water. In the field two samples were taken at each place, one with the water sample and the other with deionized water used as a field blank. The samples were filtered under a laminar flow hood and acidified with ultrapure  $HNO_3$ .

### Measurement techniques

#### Radon

The water samples were analysed for solubilized <sup>222</sup>Rn with a Packard TRI-CARB 2700TR scintillation detection system. The results are reported in Bq L<sup>-1</sup>. Corrections for decay of radon and decay and growth of the daughter products in the samples were necessary (Olguin *et al.*, 1993).

#### Physical and chemical parameters

In the field, electrical conductivity was determined with a conductimeter (Conductronic PC18). Temperature and pH were determined with a Schott pH-Meter CG 837, calibrated before each measurement using a pH 4 buffer solution. Dissolved oxygen was also measured *in situ* using an YSI Model 57 oximeter.

#### Chemical analyses

Major elements were analysed by standard methods, as given in APHA-AWWA-WPCF (1995). The accuracy of the analyses was checked by the ionic charge balance (lower than 10 % difference).

## Trace elements

Trace elements were determined at  $\mu$ g L<sup>-1</sup> levels using an ICP-MS (Inductively Coupled Plasma, Mass Spectrometer) VG Plasma Quad 2 Turbo Plus. Calibration was performed with 5 and 10  $\mu$ g L<sup>-1</sup> solution containing all the elements to be analysed. A 10  $\mu$ g L<sup>-1</sup> <sup>115</sup>In and <sup>209</sup>Bi solution was used as internal standard in order to correct instrumental drift (Morton *et al.*, 1996).

#### Bacteriological analysis

Samples for determination of total and faecal coliforms were taken in sterilised glass bottles and determined by the standard total coliform fermentation techniques at  $35 \pm 0.5$ 



Fig. 1. Location of the sampling sites around Cuitzeo lake.

and 44.5  $\pm$  0.5 °C respectively, during 24 hours (APHA-AWWA-WPCF, 1995).

## **RESULTS AND DISCUSSION**

The <sup>222</sup>Rn concentration obtained at the 7 sampling sites is shown in Figure 2. Average radon values were relatively low ranging from 0.88 to 3.66 Bq L<sup>-1</sup> indicating that the rocks have a low content of <sup>226</sup>Ra and/or a rapid transit from recharge to the output of the springs. The higher values correspond to Cuitzeo 2 in a local geological region with monogenetic volcanism composed of pyroclastic flow deposits from Cuitzeo lake. The lowest radon content corresponds to El Salitre and Copandaro having continental recent superficial deposits of alluvial and lacustrine origin. The studied



Fig. 2. <sup>222</sup>Rn concentration obtained at the 7 sampling sites.

## Table 1

Major elements (mg L<sup>-1</sup>) in the water samples from the 7 sites. The temperature T (°C), electrical conductivity E.C. (μS cm<sup>-1</sup>), total alkalinity T.A. (mg L<sup>-1</sup> CaCO<sub>3</sub>) and the ionic balance (%) are also shown.

Sample	Date	Т	E.C.	pН	T.A.	CO <sub>3</sub> <sup>2-</sup>	HCO <sub>3</sub> .	Cŀ	SO <sub>4</sub> <sup>2-</sup>	F-	SiO <sub>2</sub>	Na⁺	<b>K</b> ⁺	Ca <sup>2+</sup>	$Mg^{2+}$	<b>Bal</b> (%)
Jeruco	270301	24	1072	8.21	345.10	3.31	414.29	52.30	99.40	0.57	72.34	130.65	17.65	58.92	45.14	8.30
	160501	26	1093	7.53	332.84	0.68	404.69	50.90	98.76	0.44	64.24	98.22	12.99	54.03	50.63	3.90
Cuitzeo 2	270301	26	667	8.35	312.42	4.11	372.79	7.95	41.28	0.26	67.38	56.19	14.05	37.45	37.67	0.50
	160501	27	670	7.58	308.34	0.70	374.75	6.85	42.04	0.30	67.53	56.37	14.00	38.36	38.22	0.63
Cuitzeo 3	270301	27	483	8.40	255.25	3.76	303.76	4.25	3.92	0.20	64.10	29.21	7.38	32.88	32.13	1.80
	160501	27	496	7.67	245.04	0.69	297.55	3.94	5.18	0.21	63.38	29.27	6.73	34.18	31.21	1.70
Copándaro	160501	26	965	7.68	263.41	0.76	319.83	108.00	47.15	0.43	69.38	68.91	9.31	74.26	31.69	-1.10
Panteón	160501	28	490	7.27	249.12	0.28	303.36	4.30	1.45	0.32	81.78	45.74	7.72	45.67	21.05	4.90
El Salitre	160501	32	947	8.01	175.60	1.07	212.06	166.50	42.73	3.38	79.91	150.70	11.53	20.63	13.70	-2.30
Los Baños	160501	36	481	8.04	142.93	0.93	172.48	58.90	16.06	2.05	46.79	99.60	1.96	11.87	0.55	-0.46

wells behave quite similarly as far as <sup>222</sup>Rn content is concerned.

The results regarding major elements are shown in Table 1. The higher values for pH, Cl<sup>-</sup>, F<sup>-</sup>, Na<sup>+</sup> and CO<sub>3</sub><sup>-2-</sup> were found at El Salitre having almost the highest temperature. Jeruco showed the higher conductivity, total alkalinity, HCO<sub>3</sub> , SO42, K+, Mg+ and the second in Na+ after El Salitre. Cuitzeo 2 and 3, had differences in major elements content. Na+, K+ and Cl<sup>-</sup> were half at Cuitzeo 3 as compared to Cuitzeo 2, which are 1 km apart and a striking one order of magnitude higher SO<sub>4</sub><sup>2</sup>-value was found at Cuitzeo 2. These results indicate some differences in the water quality of these two very close boreholes. The type of water is shown in the Piper diagram (Figure 3) indicating differences between the boreholes: Jeruco and Cuitzeo 2 are of Na<sup>+</sup>-Mg<sup>+</sup>-HCO<sub>3</sub><sup>-</sup>type; Cuitzeo 3, Mg<sup>2+</sup>-Ca<sup>2+</sup>-HCO<sub>3</sub> type; Copandaro and Panteón, Ca<sup>2+</sup>-Na<sup>+</sup>-HCO<sub>3</sub>; Los Baños Na<sup>+</sup>-HCO<sub>3</sub> and finally, a quite different one El Salitre of Na<sup>+</sup>-Cl<sup>-</sup> type.

Adams *et al.*, (2001) reported the behaviour of prevailing  $Ca^{2+}$ -HCO<sub>3</sub><sup>-</sup> type of waters in higher lying areas while in topographycal flat areas Na<sup>+</sup>-Cl<sup>-</sup> type waters dominated. In the case of Cuitzeo, Israde-Alcántara (1999) explains a connection on the SW part of the Cuitzeo lake with the Zacapú zone, through a channel, indicating that El Salitre

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position has the lowest topography of the Cuitzeo zone. It has also been reported by Marini *et al.* (2001) that  $Ca^{2+}$ -HCO<sub>3</sub><sup>-</sup> facies coupled with low salinity are typical of first stages of interaction between meteoric waters and rocks. This interpretation together with the low radon concentration values could support a rapid transit of the water recharge to the discharge zone.

Trace elements (Table 2) indicate differences between the sampling sites. El Salitre had one or two orders of magnitude higher B and Mn content as compared to all the other sites. The differences between Cuitzeo 2 and 3 are also evident.

Examples of bimodal diagrams Li-B, Rb-Sr and Sr-Ba (Figure 4) indicate that each well and spring differentiates even for several samplings. Los Baños and El Salitre, located at the SW of the lake have low Sr content as compared with Jeruco (NE shore) with high Rb-Sr values. The presence of andesitic and basaltic lavas and ignimbrites at Jeruco seems to be a factor for these differences between the N and S shores of the lake. Extreme values of Ba-Sr and Rb-Sr correspond to Los Baños for the lower values and Jeruco for the largers. Los Baños, having the higher temperature, has also high F and Na<sup>+</sup> and the lowest total alkalinity. The B-Li diagram indicates that Jeruco, Panteón, Copándaro, Cuitzeo 2 and Cuitzeo 3 had similar B values with differ-



Fig. 3. Piper diagram of the sampled waters.

## Table 2

Trace elements ( $\mu g L^{-1}$ ) in the water samples from the 7 sites.

Sample	Date	Li	В	Mn	Со	Ni	Cu	Zn	As	Rb	Sr	Мо	Cs	Ba	Pb	U
Jeruco	270301	124.080	324.110	0.299	0.243	1.153	3.544	12.086	5.973	22.951	812.480	1.003	0.211	122.550	0.267	14.952
	160501	103.750	313.500	0.208	0.159	0.679	1.134	5.486	2.529	22.325	794.840	0.876	0.220	120.330	0.161	14.624
Cuitzeo 2	270301	12.801	214.330	0.031	0.193	1.413	2.777	7.374	1.914	20.573	500.630	1.462	0.385	49.255	0.663	8.001
	160501	12.116	204.250	0.214	0.102	0.544	1.202	9.425	0.958	20.664	511.050	1.678	0.389	48.876	0.165	8.084
Cuitzeo 3	270301	7.413	51.963	0.136	0.128		1.726	4.464	1.027	12.051	341.190	0.525	0.728	28.944	0.270	3.121
	160501	7.009	51.679	0.187	0.081	0.508	2.302	10.827	0.471	12.047	344.940	0.609	0.739	27.459	0.331	3.072
Copándaro	160501	45.069	186.830	0.390	0.206	1.153	1.309	2.599	7.501	11.644	764.440	2.234	0.108	59.715	0.114	4.302
Panteón	160501	52.122	53.403	0.643	0.122	0.806	24.206	7.490	2.580	13.215	325.940	0.742	0.094	26.611	1.871	2.564
El Salitre	160501	105.190	2192.700	5.299	0.058	0.539	1.169	4.045	8.290	22.613	218.190	4.649	1.556	86.284	0.162	1.740
Los Baños	160501	129.320	991.890	0.251	0.034	0.090	0.919	1.696	16.851	4.950	35.291	3.400	5.073	1.489	0.098	0.864



Fig. 4. Examples of bimodal diagrams Li-B, Rb-Sr and Sr-Ba.

ences in Li, almost one order of magnitude between the extremes Jeruco and Cuitzeo 3. El Salitre had the maximum B, one order of magnitude higher than all the other samples. The temperature, water type, high boron and lithium contents suggest that El Salitre and Los Baños are part of the same system.

No microbiological contamination was found in the studied samples.

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## BIBLIOGRAPHY

- ADAMS, S., R. TITUS, K. PIETERSEN, G. TREDOX and C. HARRIS, 2001. Hydrochemical characteristics of aquifers near Sutherland in the Western Karoo, South Africa. J. Hydrol. 241, 91-103.
- ANNANMAEKI, M., T. TURTIAINENE, H. JUNGCLAS and C. RAUSSE, 2000. Disposal of radioactive waste arising from water treatments: recommendations for the EC. Final Report of the WP8 of the TENAWA Project. Radiation and Nuclear Safety Authority, Helsinki, Finland.
- APHA-AWWA-WPCF, 1995. Methods for the examination of water. 19<sup>th</sup> edition, American Public Health Association (APHA), American Water Works Association (AWWA), Water Pollution Control Federation (WPCF), Washington D.C.
- CORBETT, C. R. and W. C. BURNETT, 1997. Radon tracing in groundwater input into Pad Pond, Sanannah River site. J. Hydrol. 203, 209-227.
- DE CSERNA, Z. and R. ÁLVAREZ, 1995. Quaternary drainage development in Central Mexico and the threat of an environmental disaster: a geological appraisal. *Environ. Engin. Geoscience*, *1*, 59-34.
- HAMADA, H., 2000. Estimation of groundwater flow rate using the decay of <sup>222</sup>Rn in a well. *J. Environ. Radiact.* 47, 1-13.
- ISRADE-ALCÁNTARA, I., 1999. Los lagos volcánicos y tectónicos de Michoacán. *In:* Carta Geológica de Michoacán. Escala 1:250 000.Corona Chávez P. and Israde-Alcántara I. Editors. Universidad Michoacana de San Nicolás de Hidalgo, Morelia, 45-73.
- KITTO, M. E. and M. K. KUHLAND, 1995. Radon measurements in groundwater. J. Radioanal. Nucl. Chem. 193, 253-258.

- MARINI, L., M. CANEPA, F. CIPOLLI, G. OTTONELLO and M. V. ZUCCOLINI, 2001. Use of stream sediments chemistry to predict trace elements chemistry of groundwater. A case study from the Bisagno valley (Genova, Italy). *J. Hydrol. 241*, 194-220.
- MORTON, B. O., M. A. ARMIENTA, E. HERNÁNDEZ and E. LOUNEJEVA, 1996. Espectrómetro de masas con Plasma de Acoplamiento Inductivo de Geofísica. UNAM. Actas INAGEG 2. México. pp 149-154.
- OLGUIN, M. T., N. SEGOVIA, E. TAMEZ, M. ALCÁNTARA and S. BULBULIAN, 1993. Radon concentration levels in groundwater from the City of Toluca, Mexico. *Sci. Tot. Environ.* 130/131, 43-50.
- PAULUS, L. R., T. F. GESELL and R. R. BREY, 1998. An evaluation of <sup>222</sup>Rn concentration in Idaho groundwater. *Health Phys.* 74, 237-241.
- SEGOVIA, N., E. TAMEZ, P. PEÑA, J. CARRILLO, E. ACOSTA, M. A. ARMIENTA and J. L. ITURBE, 1999. Groundwater flow system in the Valley of Toluca, Mexico: an assay of natural radionuclide specific activities. *Appl. Radiat. Isotop. 50*, 589-598.

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