# Radon in soil and chemical composition of spring water near Popocatépetl volcano, Mexico

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#### RESUMEN

Se determinaron fluctuaciones de radón en suelo en dos estaciones ubicadas en el flanco norte del volcán Popocatépetl. Así mismo se midió radón y se determinó la composición química de muestras del agua de tres manantiales localizados alrededor del volcán. Las medidas de radón en suelo se realizaron con detectores sólidos de trazas nucleares y ocasionalmente con registros en periodos cortos con sondas Clipperton. El radón en agua se midió por el método de centelleo líquido. Las componentes químicas mayores del agua se cuantificaron por métodos químicos convencionales y los elementos traza por medio de un ICP-MS. Los niveles del radón en suelo mostraron patrones de comportamiento distinto durante 1999–2000. Los compuestos químicos del agua indican diferencias en el origen de cada manantial.

PALABRAS CLAVE: Radón, suelo, agua subterránea, composición química, volcán Popocatépetl, México.

#### ABSTRACT

Soil radon was monitored at two permanent stations on the northern flank of Popocatépetl volcano. Water samples from three springs around the cone were also studied for radon and chemical composition. Radon in soil was recorded using track detectors, and sporadic short-term measurements were obtained with a Clipperton probe. Radon in water samples was measured using a liquid scintillation method. The chemical composition was obtained from conventional chemical methods and trace elements using an ICP-MS equipment. Soil radon levels obtained with track detectors at the two monitoring stations showed different patterns in 1999-2000. The chemical composition indicates differences in the origin of the springs.

KEYWORDS: Radon, soil, groundwater, chemical composition, Popocatépetl volcano, Mexico.

### **INTRODUCTION**

Radon as a tracer of pore fluids is useful in the study of volcanic eruptions. Long-term soil gas radon surveys have been performed in order to observe possible fluctuations in active volcanoes (De la Cruz Reyna *et al.*, 1985; Segovia and Mena, 1999; Segovia *et al.*, 2001). However, medium (Connor *et al.*, 1996) and short-term relationships of soil radon using high time-resolution probes showed that radon anomalies may be correlated with a sudden eruptive crisis (Seidel *et al.*, 1999), or with fault systems in active volcanoes (Varley and Armienta, 1999). Radon and chemical composition of groundwater from springs and wells on the mountain side of active volcanoes have also shown changes with eruptions (Notsu *et al.*, 1983; Segovia *et al.*, 1999; Armienta and De la Cruz-Reyna, 1995).

Since 1994, Popocatépetl volcano started an eruptive phase. Its geographical position, about 60 km from Mexico City, with the largest population concentration in the country, and the evidence of large eruptions in historical times (Siebe *et al.*, 1996) makes this a high - risk volcano. The recent activity has been moderate, mostly restricted to periodic emissions of fine ashes and gas.

Measurements of soil radon and groundwater chemistry have been performed at different sites around the volcano during several years (Segovia *et al.*, 1997, 1999, 2001), indicating slight changes in the studied parameters as a function of increased activity of the volcano. The volcanic monitoring continues, due to sporadic eruptions. The main purpose of the present paper is to analyse the effect of recent volcanic activity on several parameters such as soil and groundwater radon and chemical composition of groundwater.

#### **EXPERIMENTAL**

The sites. Popocatépetl volcano (5465 m) is located in the central part of the Mexican Neo-Volcanic Belt

(19°03'36"N; 98°31'18"W). In December 1994, an eruptive stage started and evolved with fluctuations over the last 6.5 years. In the period 1999-2001 the volcanic explosivity index (VEI) was lower than one (Newhall and Self, 1982). However, in December 2000, an eruption occurred and the population living around the volcano had to be evacuated.

The soil gas radon monitoring was performed at two fixed stations located at 7.5 and 4.7 km from the crater at Paso de Cortés (3400 m) and Tlamacas (4000 m) on the northern flank of the Popocatepetl volcano. These two stations are 5 km from each other and their soil characteristics are quite different due essentially to the altitude factor: Paso de Cortés station corresponds to a conifer and oak forest with the top soil organic matter layer up to almost 70 cm, while Tlamacas is found already at the upper cinder cone of the volcano above the tree line, with the top soil formed from volcanic ashes having no vegetation.

Groundwater radon was measured in water samples obtained from the two springs, Atlimeyaya and Axocopan, located on the eastern part of the volcano. Some measurements were also performed at Calvario spring in the southern mountain side (Figure 1). The geology associated with the springs indicates andesitic and basaltic rocks. The spring waters are used mainly for domestic activities such as washing and local drinking water supply; a fish hatchery is located near Atlimeyaya.

*Water sampling.* Monthly water sampling for radon determination was carried out. The methodology has been previously reported (Olguin *et al.*, 1993).

One litre water samples were taken in plastic polyethylene bottles for major chemical components such as Cl<sup>-</sup>, SO<sup>2-</sup><sub>4</sub>, and SiO<sub>2</sub>. A 500 ml sample with 3 ml HNO<sub>3</sub> added was used for the determination 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 with 60 ml polyethylene bottles previously washed and decontaminated. One day before the sampling the bottles and covers were rinsed 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<sub>3</sub> (1% v/v).



Fig. 1. Location of the soil monitoring sites at Paso de Cortés and Tlamacas and the three water springs: Axocopan, Atlimeyaya and Calvario.

## Measurement techniques

<u>Radon.</u> Long term soil radon determination (70 cm depth) was performed with Solid State Nuclear Track Detectors (SSNTD), LR-115 type II, from Dosirad Co., France. Sporadic records of soil radon in the short term were performed using two Clipperton probes at Paso de Cortés station. The details of the methodology have been previously reported (Segovia *et al.*, 1997, 2001).

The water samples were analysed for dissolved radon with a Packard TRI-CARB 2700TR liquid scintillation detection system (Olguin *et al.*, 1993).

*Physical and chemical parameters of the water samples.* 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 4 pH buffer solution.

Chemical analyses were performed by standard methods, as given in APHA-AWWA-WPCF (1995). The accuracy of the analyses was checked by the ionic charge balance (lower than 4% difference).

Trace elements were determined at  $\mu$ g L<sup>-1</sup> levels using an ICP-MS (Inductively Coupled Mass Spectrometer) VG Plasma Quad 2 Turbo Plus at the University of Montpellier, France. The samples are delivered through international postal service. 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.*, 1998).

## **RESULTS AND DISCUSSION**

The soil radon fluctuations observed at Paso de Cortés and Tlamacas with SSNTDs in 1999-2000 are shown in Figure 2a. The average radon in soil values were quite low and the average radon concentration values were about twice at Paso de Cortés as compared with Tlamacas. This behaviour was observed in previous years (Segovia et al., 2001). However it is noticeable that the radon concentration values at Tlamacas, the closest station to the cone, increased in the middle part of 2000 as compared with 1999, reaching the maximum in December 2000, when the eruption occurred. However, the behaviour of soil radon at Paso de Cortés had the same pattern in 1999 and 2000, but with higher concentration values of soil radon during the rainy season of 2000. It has been reported (Segovia et al., 2001) that, during the period 1997-1999, a higher number of anomalous soil radon values (higher than  $\overline{x}+2\sigma$ ) occurred at Tlamacas station as

compared with Paso de Cortés, when measuring with SSNTDs. This behaviour has been explained as related to the increasing seismicity due to volcanic events that generate differences in the underground gas flow related to the type of soil.

An example of short term monitoring is shown in Figure 2b. The records correspond to two probes located nearby (1 m from each other, having independent energy supply systems) at Paso de Cortés station. The response of both equipment's was very similar and showed a peak on 14 April, 2001, at 17:38 and 17:56 respectively, two days before an eruption taking place on April 16, 2001, at 19:45 which sent volcanic bombs 2 km away from the crater and produced an ash plume that rose 4 km. The time difference in the response of the two equipments is probably due to local inhomogeneities at the top layer of the soil. However, a certain concordance in the response of the two equipments to a same radon signal is observed.

Groundwater radon at the springs around the volcano (Figure 3) indicated a seasonal behaviour with a noticeable increase in May-June 2000 corresponding to a maximum in the precipitation of this special year. The lowest radon data occurred at the end of the rainy season in September-October. The three springs had almost the same radon behaviour during the year and fluctuations were mostly correlated with seasonal changes.

Stability was observed for chemical species and trace elements at Axocopan and Atlimeyaya springs for different samplings, performed from 1997 to 2000. An example of the values obtained is shown in Table 1. The chemical species had higher concentration values at Axocopan. Trace elements such as Li, B, Cr, Rb, Sr, Mo, Ba and U had also two or three times higher values at Axocopan than Atlimeyaya in the water samples, indicating that the first spring had greater mineralization characteristics.

The correlation matrix of the available data of trace elements at Atlimeyaya and Axocopan is shown in Table 2, indicating strong positive correlation between pair of elements.

Examples of bimodal diagrams Rb-Sr, B-Li, Ba-Sr and Li-Sr (Figure 4) indicate the differentiation of the two springs, showing peculiarities in their water origin.

For the studied eruptive period, the soil radon fluctuations indicate a certain effect that reflects volcanic activity changes. The water chemistry of nearby springs was stable in the time, even if the water characteristics showed differences in their origin. The present results agree with the low volcanic activity from the Popocatépetl in the period of study.





(b)



Fig. 2. (a) Soil radon at Tlamacas and Paso de Cortés during 1999-2000. (b) Radon signals from the Clipperton instrument. The peak occurred 2 days before the eruption (arrow).



Fig. 3. Radon concentration (Bq L<sup>-1</sup>) in the water samples at Atlimeyaya, Axocopan and Calvario during 2000-2001.

## Table 1

Examples of major species (mg L<sup>-1</sup>) and trace elements (µg L<sup>-1</sup>) in spring waters from Atlimeyaya and Axocopan

Data	Site	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na⁺	K⁺	Cl	SO42-	HCO3-	total alk.	F <sup>-</sup>	SiO <sub>2</sub>	T (°C)	pН	E. C. (µS cm <sup>-1</sup> )	Bal
3/07/2001	Axocopan	35.4	42.3	47.3	6.6	15.0	38.8	398.7	326.8	0.5	83.7	20.0	6.1	589	-1.8
3/07/2001	Atlimeyaya	12.8	7.1	12.8	2.8	5.6	7.5	98.1	80.4	0.5	54.3	14.0	6.5	143.0	-1.7

## **MAJOR SPECIES**

#### TRACE ELEMENTS

Data	Site	Li	В	Cr	Mn	Со	Cu	Zn	As	Rb	Sr	Мо	Cd	Cs	Ва	Pb	U
30/3/00	Axocopan	75.96	256.05	3.88	0.04	0.08	0.34	5.63	1.23	15.87	239.06	3.66	0.05	0.42	22.81	0.10	1.06
30/3/00	Atlimeyaya	17.35	117.13	0.59	0.07	0.03	0.49	4.84	1.12	7.20	52.28	1.45	0.01	0.33	5.29	0.06	0.20

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Fig. 4. Bimodal diagrams for Rb-Sr, Li-Sr, Li-B and Sr-Ba.

Table	2
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Trace elements correlation matrix for the Atlimeyaya and Axocopan water samples

Corr	elation	matrix														
	Li	В	Cr	Mn	Со	Cu	Zn	As	Rb	Sr	Mo	Cd	Sc	Ba	Pb	U
Li	1.000															
В	0.998	1.000														
Cr	0.996	0.999	1.000													
Mn	0.442	0.404	0.413	1.000												
Co	0.987	0.981	0.982	0.504	1.000											
Cu	0.078	0.122	0.148	-0.401	0.123	1.000										
Zn	0.585	0.634	0.650	-0.168	0.539	0.677	1.000									
As	0.732	0.727	0.718	0.473	0.814	0.255	0.333	1.000								
Rb	0.999	0.996	0.992	0.447	0.990	0.069	0.563	0.752	1.000							
Sr	1.000	0.996	0.994	0.463	0.989	0.059	0.563	0.741	0.999	1.000		_				
Mo	1.000	0.998	0.995	0.423	0.987	0.088	0.590	0.741	0.999	0.999	1.000		_			
Cd	0.439	0.410	0.371	0.522	0.461	-0.555	-0.258	0.607	0.467	0.459	0.447	1.000				
Sc	0.963	0.952	0.943	0.519	0.980	0.007	0.420	0.856	0.973	0.969	0.966	0.621	1.000			
Ba	0.999	0.995	0.993	0.477	0.988	0.046	0.558	0.730	0.998	1.000	0.998	0.455	0.966	1.000		
Pb	0.378	0.336	0.342	0.979	0.465	-0.358	-0.256	0.545	0.389	0.401	0.362	0.568	0.501	0.412	1.000	
U	0.998	0.995	0.995	0.485	0.989	0.066	0.573	0.730	0.997	0.999	0.996	0.434	0.961	0.999	0.418	1.000

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