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Radon and hydrogeochemical monitoring at Popocatépetl volcano, Mexico

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RESUMEN
Se efectuaron determinaciones de radón en suelos alrededor del volcán Popocatépetl de septiembre 1997 a abril 1999. El gas del suelo se muestreó con un tubo de acero inoxidable y se midió en el campo con un contador de centelleo de partículas alfa. En uno de los sitios se correlacionaron las variaciones de las concentraciones de 222Rn con los cambios en la actividad volcánica. Por otro lado, se sabe que las variaciones en las características químicas de manantiales en volcanes activos pueden utilizarse para el monitoreo volcánico. En 1993 inició un período de actividad del volcán Popocatépetl. Desde 1995, el análisis químico de manantiales ha sido incluido dentro de la vigilancia volcánica. En el campo se midieron la conductancia, el pH y la temperatura. Las concentraciones de iones principales HCO3-, Cl-, SO4²-, Mg²+, Na⁺, K⁺, Ca²⁺ se determinaron en el laboratorio por métodos estándares. Las especies menores boro, sulfuro y fluoro se analizaron por colorimetría y potenciometría. Durante el período de estudio se observaron incrementos de las concentraciones de B, SO₄²⁻ y Cl⁻ antes de algunos cambios en la actividad, en algunos manantiales. Se encontró que las anomalías de radón coincidieron aproximadamente con algunas de las variaciones químicas. Los niveles más altos de ²²²Rn en uno de los sitios se obtuvieron en el período que precedió a la erupción del 24 de diciembre de 1997, la cual incluyó la emisión de fragmentos de tefra relativamente grandes.

PALABRAS CLAVE: Radón, Popocatépetl, monitoreo geoquímico, manantiales, suelos.

ABSTRACT
Measurements of radon in soil gas were carried out around Popocatépetl volcano during the period from September 1997 to April 1999. Soil gas was sampled using a stainless steel probe, and measured in the field with an alpha scintillation counter. At one location concentration variations of ²²²Rn in soil have been related with changes in the volcanic activity. In addition, chemical variations of spring waters at active volcanoes may be useful for volcano monitoring. In 1993 an increased period of volcanic activity began at Popocatépetl volcano. Since 1995, the chemical analyses of springs have been carried out as part of the surveillance of the volcanic activity. Conductivity, pH, and temperature were measured in the field. Concentration of the major ions: HCO₃⁻, Cl⁻, SO₄²⁻, Mg²⁺, Na⁺, K⁺, Ca²⁺ have been determined in the laboratory by standard methods. The minor species boron, sulfide and fluoride were analyzed by colorimetry and potentiometry. Concentration increases of B, SO₄²⁻ and Cl⁻ have been observed preceding some changes in volcanic activity during the studied period in some of the springs. Radon anomalies were found to approximately coincide with some of the chemical variations. The highest ²²²Rn levels at one location were measured during the period that preceded the eruption that occurred on December 24, 1997, which included the emission of relatively large tephra fragments.

KEY WORDS: Radon, Popocatépetl, geochemical monitoring, springs, soils.

INTRODUCTION
Volcanic risk assessment is of paramount importance when millions of people are exposed to volcanic activity. Different methods must be simultaneously applied to assess the changes in the volcanic system that may result in a dangerous eruption. Geochemical monitoring methods have been proved to be able to detect activity changes in different volcanoes (Giggenbach, 1983; De la Cruz-Reyna et al., 1989; Armienta and De la Cruz-Reyna, 1995; Martini,1996; Armienta et al., 2000; Taran et al., 2000). Popocatépetl is an active volcano located at 19°01.38' and 98°37.29, within the Mexican Volcanic Belt. It is an andesitic-dacitic stratovolcano formed by a succession of deposits from different eruptive stages. The summit of Popocatépetl volcano is the second highest peak of Mexico with a height of 5454 meters above sea level, and covers an area of about 500 km². The location of Popocatépetl volcano near three important cit-
ies (65 km southeast of Mexico City, 42 km southwest of the city of Puebla and 65 km northeast of the city of Cuernavaca) makes it a potential risk.

The current stage of Popocatépetl eruptive activity began in early 1993 with a considerable increase in seismicity and gas emissions that culminated with an explosion sequence on December 21, 1994. The activity which has been characterized by gas and tephra emissions, and the growth and destruction of lava domes, continues until present.

In strato-volcanoes, meteoric waters percolate through the various layers that form the strata system resulting from the various eruptive stages. At Popocatépetl volcano, the layers are formed by tuff sequences, ashes, pumice and various intercalated lava layers of different types. These rock layers exhibit basic to intermediate and acid compositions. At certain points mudflows and avalanche deposits form part of this sequence. Additionally, there are faults and cracks that allow more rapid water percolation and volcanic gas transport. The aquifers within or near the volcanic edifice may interact with volcanic gases and along with water-rock interactions its chemical composition can be modified. Changes in the level of volcanic activity have been reflected by variations in the chemistry of springs and wells at various volcanoes (Takahashi et al., 1988; Gislason et al., 1992; Schevenell and Goff, 1995; Armienta and De la Cruz-Reyna, 1995; Tilling and Jones, 1996). In addition, the radon content of soil gas has been used to detect faults in a volcanic edifice (Ball et al., 1991), to estimate the volume of degassed magma (De la Cruz-Reyna et al., 1985), and as an indicator of volcanic process evolution (Connor et al., 1996; Segovia et al., 2001).

The aim of this work was to study the radon concentration variations in soils and the chemical characteristics of springs in relation to Popocatépetl activity, from September 1997 to December 1999.

METHODS

Water sampling was done in five springs located southwest of the volcanic edifice. At each sampled spring temperature, conductivity and pH were measured in situ with a conductimeter Conductronic PC18, calibrated with pH buffers (4.0, 7.0 and 9.0). Three water samples were taken in plastic containers of 1000, 500 and 125 ml, and preserved with different chemicals. Chemical analysis was performed at the Geophysics Institute, UNAM. The concentration of the major ions: HCO₃, Cl⁻, SO₄²⁻, Mg²⁺, Na⁺, K⁺, Ca²⁺, was determined by titration, turbidimetry, potentiometry with selective electrodes, and emission spectroscopy following standard methods of water analyses (APHA, 1995). The minor species boron and sulfide were analyzed by colorimetry, and fluoride was measured by potentiometry.

Radon in soil gas was sampled using a stainless steel probe, at a depth of about 0.6 m. ²²²Rn was measured in situ with an alpha scintillation counter. More than 500 measurements were made along various traverses on the volcano flanks to try and locate spatial anomalies (Varley and Armienta, 2001). Latitude and longitude were obtained with a handheld GPS at each sampled point. At one location, in a ravine northwest of Popocatépetl (Palo Rechino), radon was measured approximately every month. Unfortunately, not all the dates were coincident for soil radon measurements and spring water sampling. Radon determinations were performed monthly from September 1997 to April 1999. Water sampling was carried out in October and December 1997, and between June 1998 and December, 1999 the frequency was almost monthly.

RESULTS AND DISCUSSION

Spring waters had acid to slight alkaline pH values, between 5.7 and 7.9 with only slight temporal changes at each spring. Nevertheless, a significant anomaly occurred at Paleorío in October 1997, when a pH value of 6.9 was measured, followed by values of 7.5 ± 0.1 throughout the sampling period. Specific temperature ranges were observed at each spring (from 7°C to 20°C) and showed small variations throughout the year. The higher temperatures measured at Axocopan and Huexocoapan may be related to their lower altitude and to a lesser influence of the Popocatépetl glacial melt. Low conductivity values were determined in four of the springs (150-250 µS/cm), a higher conductivity was measured at Axocopan (700 µS/cm). All of the springs are mixed bicarbonate type waters. Concentration of the main ions did not show strong temporal variations. Nevertheless, certain chemical species had small changes on certain dates. Two sulfate concentration peaks were observed, the first one in Huexocoapan in September, 1998, and the second one in Axocopan on March 3, 1999 (Figure 1). Boron was below the detection limit (0.5 mg/L) most of the period. However, an increase was observed at Axocopan, Paleorío and Teconala in December 1998 and also at Atlimeyaya in January 1999. Another increase was measured in May and August that year, but only in Atlimeyaya and Axocopan. In October a boron peak appeared in all the springs (Figure 2). Concentration variations of chloride followed approximately the same pattern at Atlimeyaya, Huexocoapan, Paleorío and Teconala. (Figure 3). A concentration peak of chloride occurred on April 1999. The spring at Axocopan behaved differently decreasing on that date; an important chloride increase was recorded from October to December 1999.

The highest ²²²Rn in soil gas values (12.47 kBq/m³ and 12.17 kBq/m³) measured in the bottom of the Palo Rechino ravine, were observed in September and November 1997 (Figure 4); lower levels were measured afterwards.
Fig. 1. Sulfate concentrations (mg/L) at the sampled springs against time.

Fig. 2. Boron concentrations (mg/L) at the sampled springs against time.
The highest $^{222}$Rn levels were observed before the appearance of a lava dome on December 6, 1997. Relatively large tephra fragments were emitted in this explosion. Measurements made through the following wet season in 1998 did not show any increase. This indicates that the anomaly was not associated with seasonal changes. This variation may be explained by tectonic stress associated with local earthquakes or increasing stress before the volcanic eruptions. These stress-state changes could have increased gas transport through fractures and faults, resulting in increased radon levels. Volcanic acid gas dissolution could have also produced the low pH at Paleorío in October, 1997.

A moderate increase of Popocatépetl activity occurred during September and October, 1998, after a stable and low-level period from May to August. This increased activity was characterized by exhalations of vapor and gas, sometimes accompanied by a low amount of ash emissions. A slight radon anomaly on August and a sulfate increase on September, 1998, may be associated with these increases in activity.

Boron and sulfate concentration peaks in January and the beginning of March, 1999 preceded the volcanic activity occurred in March 1999. Since the beginning of that month periods of low-frequency harmonic tremor were registered followed by eruptions that emitted rock fragments near the summit, and ashes that reached the city of Puebla on March 12. An explosion was registered in the morning of April 15, 1999; a chloride concentration peak was measured the same day. The boron peaks of May and August, 1999 were not followed by a volcanic activity increase, since from May to December, 1999 the overall activity level remained low, with the occurrence of only small sporadic ash emissions.

**CONCLUSIONS**

Concentration increases of B, $\text{SO}_4^{2-}$, Cl have been observed preceding some of the changes in the volcanic activity during the studied period. Anomalies were observed simultaneously in all the springs only on certain dates. $^{222}$Rn anomalies were found to approximately coincide with some of the chemical variations. The highest $^{222}$Rn levels were measured during the period that preceded the extrusion of lava and tephra emission in December 1997. Increased fracturing and enhanced fluid flow resulting from volcanic activity at Popocatépetl, may produce higher soil radon levels and greater concentrations of ions linked to volatile emissions in waters. Geochemical monitoring of springs and regular measurement of $^{222}$Rn in soils may provide important information to assess the risk at Popocatépetl volcano. Shorter sampling periods are necessary to improve the interpretation.

![Fig. 3. Chloride concentrations (mg/L) at the sampled springs against time.](image-url)
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