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Experimental studies of anomalous radon activity in the Tlamacas Mountain, Popocatepetl Volcano area, México: new tools to study lithosphere-atmosphere coupling for forecasting volcanic and seismic events

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ABSTRACT

This study presents and discusses the results of soil radon monitoring at three different volcano sites and one reference site, from December 2007 to January 2009. This relates to the activity of the Popocatepetl Volcano and a radon survey and gamma-ray spectrometry in the area between Paso de Cortes and Tlamacas Mountain, and in the adjacent regions. The results are applied to the aspects of atmosphere electricity and lithosphere-atmosphere coupling in relation to the forecasting of volcano and earthquake activity. The monitoring of radon release reveals a decrease in radon concentration (down to total suppression) with approaching moderate volcanic eruptions. The behavior of the radon activity at the Tlamacas site is more apparent, compared to other observational sites. The average level of radon release observed at the Tlamacas site is much higher, with some characteristic variations. Both the radon survey and gamma-ray spectrometry indicate intensive diffusion radon emission localized in the area of Tlamacas Mountain. The average radon concentration in the area of Tlamacas is about 10-20-fold greater than the background volcano values. The new concept of lithosphere-atmosphere coupling is presented: intensive radon release in high elevated areas shortens and modifies the Earth-to-thunderclouds electric circuit, which provokes micro-discharges into the air close to the ground, attracting lightning discharges. This concept attempts to explain in a new way the noise-like geomagnetic emissions registered before major earthquakes, and it promotes interest for the study of thunderstorm activity in seismo-active zones, as a promising instrument for earthquake forecasting.

1. Introduction

The Popocatepetl Volcano (nicknamed «El Popo») is located in Central Mexico (latitude, 19.07°N; longitude,

98.63°W; altitude, 5,465 m). It is one of the active volcanoes that forms the Trans-Volcanic Belt of Mexico (also known as the Neo-Volcanic Axis), and its existence is related to the geodynamics of the North American and Coco plates. El Popo is a major potential hazard in Mexico, and therefore its eruptive activity is a subject of permanent monitoring by the National Center for Prevention of Disasters (CENAPRED). Every day, a summary of the behavior of Popocatepetl Volcano can be found on the CENAPRED webpage (<http://www.cenapred.unam.mx/cgi-bin/popo/reportes/consultai.cgi>).

The activity of Popocatepetl Volcano can be briefly summarized as follows. Light eruptions are permanent everyday events, with several to tens of eruptions of gas and water seen daily during the quiet volcano phases, and up to 50-100 eruptions occurring in the active phases [see González-Pomposo 2004, and Table 2 in Arámbula-Mendoza et al. 2010, for detailed description of the recent Popocatepetl activity stages]. Moderate eruptions of gases and volcano ash take place from once every few months up to several times per day, for the quiet and active phases, respectively. Moderate eruptions can also be accompanied by explosive elements (rock ejection), with the frequency of such events as several to dozens of events per year in recent times. Major eruptive activity is not so frequent, and the last time it occurred it lasted from December 2000 to January 2001. At this stage, intensive rock expulsions, lava eruptions and pyroclastic flows can take place, in addition to the above-mentioned phenomena. Tectono-volcanic micro-seismic events with magnitudes up to Ms 2-3 and high frequency tremors are also part of the volcano activity.

Our earlier studies in the Popocatepetl Volcano area (2003-2006) showed a variety of geomagnetic anomalies that were associated with volcano activity [Kotsarenko et al. 2007] and also allowed us to propose the hypothesis of the existence of a second magmatic reservoir [Kotsarenko et al. 2008]. The present study is devoted to various aspects of radon monitoring that was carried out in the first stage of our experimental investigations into the area of Popocatepetl Volcano (December 2007 to December 2009).

In the first part of this study, we present the results of the soil radon monitoring that was carried out at three different sites in the volcano area, and we discuss the phenomena that accompany volcano eruptions. Similar studies have been carried out in different active volcanoes all over the world, as a perspective tool for the forecasting of volcano hazard, including in Italy [Baurbon et al. 1991, Cigolini et al. 2001, 2005], France [Segovia et al. 1997, Toutain et al. 2002], Spain [Viñas et al. 2007], the Azores [Aumento 2002], Colombia [Londoño 2009], Costa Rica [Barquero et al. 2005, García-Vindas et al. 2002], Mexico [Armienta et al. 2002, Segovia 1991, Segovia et al. 1997, 1999, 2001, 2003, 2005, 2007, Varley and Armienta 2001, Varley 2003, Varley and Taran 2003], Nicaragua [Connor et al. 1996], and Indonesia [Baurbon et al. 1991].

The second part of this study deals with combined studies using radon surveys and gamma-ray spectrometry. Gamma rays are the most penetrating radiation from natural and man-made sources, so gamma-ray spectrometry is a powerful tool for monitoring and assessing the radiation

environment [IAEA 1989, 2003]. Potassium, uranium and thorium are sensitive to tectonic fractionation, so that gamma-ray spectrometry provides a method of conveniently and rapidly measuring compositional changes over large areas of rough terrain [Thorpe 1995, Chiozzi et al. 1998]. Radon surveys are recognized as a reliable tool for revealing tectonic structures [King 1980, King et al. 1993, 1994, 1996, Chyi et al. 2002, Etiope et al. 2005, Vivek et al. 2005] and volcano-tectonic geological formations [Martín et al. 2003, Burton et al. 2004, Hernandez et al. 2004].

In the third part of the present study, we examine previously observed geomagnetic anomalies [Kotsarenko et al. 2007] and enhanced thunderstorm activity, in terms of the lithosphere-atmosphere coupling caused by intensive radon release under specific conditions in the area of Tlamacas Mountain.

2. Soil radon monitoring in the Popocatepetl Volcano area

2.1. Equipment and methods

Portable solid-state shallow subsurface radar (SHARAD) scout detectors were used for the monitoring of radon concentrations in the soil at the Tlamacas station (latitude, 19.065°N; longitude, 98.63°W; referred to as «Tlamacas»), at a site near Paso de Cortes (latitude, 19.07°N; longitude, 98.65°W; «Paso de Cortes»), at a site near Tlamacas Mountain (latitude, 19.07°N; longitude, 98.63°W; «Tlamacas 2»), and at an Amecameca referent site (latitude, 19.12°N; longitude, 98.77°W) (Figure 1). These instruments use the diffusion

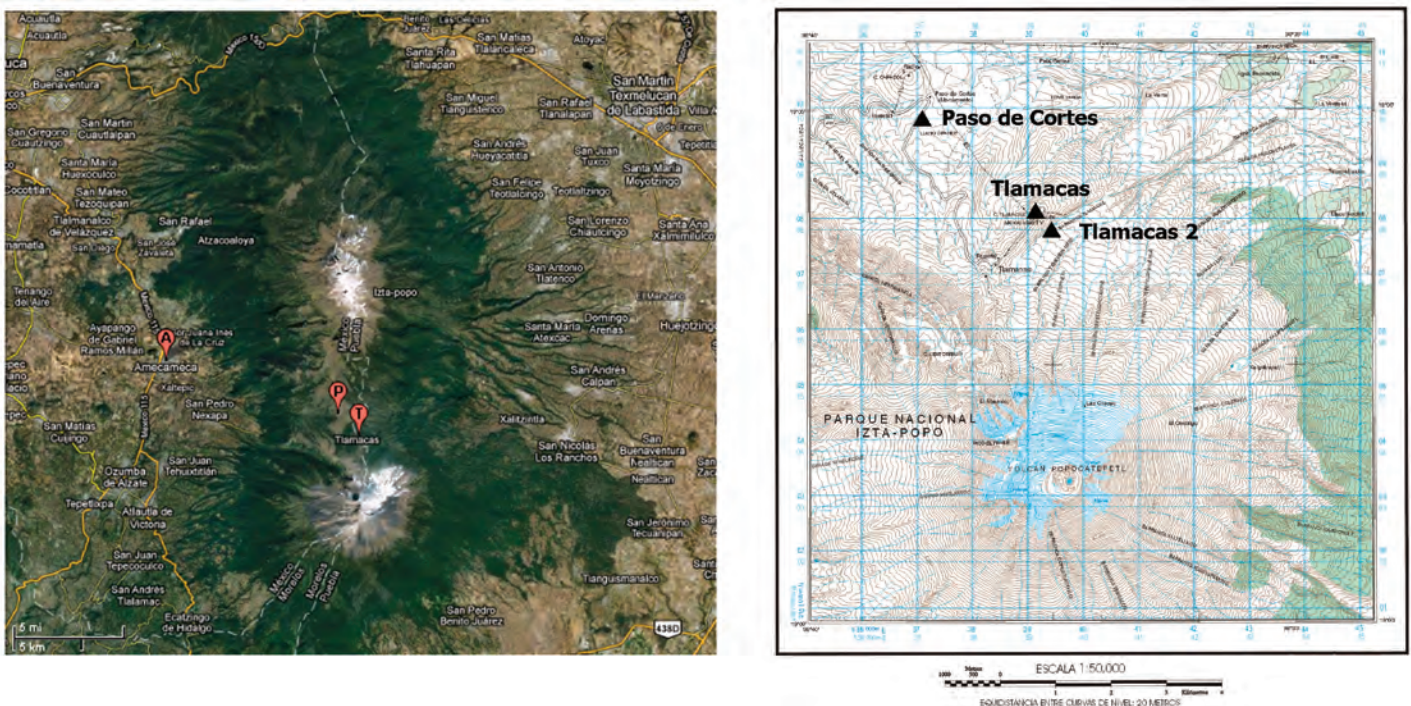


Figure 1. Left: Map showing the Amecameca referent site (A), and the Paso de Cortes (P) and Tlamacas (T) Sites. Right: Map of volcanic area showing the Paso de Cortes, Tlamacas and Tlamacas 2 sites.

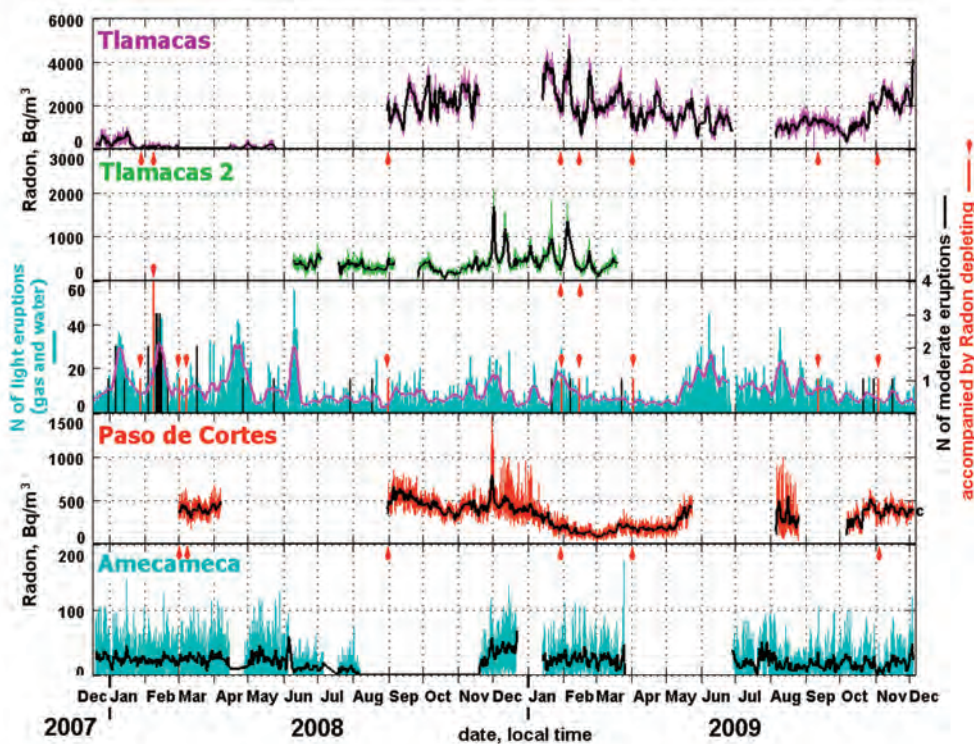


Figure 2. Radon emission monitoring at the three volcano sites. Top to bottom: Tlamacas, Tlamacas 2, Paso de Cortes and Amecameca referent site. Monitoring from December 2007 to December 2009. Middle panel: Volcano eruptions. Blue bars, eruptions of gas and water; black bars, moderate eruptions; red bars and arrows, moderate eruptions where radon decreases were observed.

method of air sampling (without pumping the air into the chamber), and therefore in a normal environment (here, «normal» meaning «not aggressive»; see Streil et al, 2008), the dominant monitored content comes from the most stable isotope of ^{222}Rn (referred to here as «radon»; $T_{1/2} = 3.8$ days; produced in the ^{238}U radioactive decay chain). Another isotope, ^{220}Rn (referred to here as «Toron»; $T_{1/2} = 55.6$ s; parent element, ^{232}Th) is rarely detectable under normal environmental conditions, as it decays before it can penetrate into the chamber. The scout detectors were also equipped with simple sensors to measure the basic meteorological parameters: temperature, relative humidity, and atmospheric pressure, in addition to radon concentration. Nine detectors were used alternatively for measuring the radon at these monitoring sites, with sampling rates of 1 point per 60 min (for instruments with internal memory, 2047 pts), and 1 point per 90 min (1023 pts), to provide uninterrupted data for each consistent époque of measurements, with a duration of 1-2 months.

2.2. Results and discussion

The monitoring of the radon emissions at these four stations (Figure 2) showed some stable tendencies. First of all, the average values of the radon concentrations observed at the Tlamacas site were normally 1.5-5-fold greater than those measured at the Tlamacas 2 site, 2-15-fold greater than at Paso de Cortes, and 25-400-fold greater than the Amecameca reference site values. Then, there was a distinct difference

between the data recorded at the volcano sites. The Paso de Cortes and Tlamacas 2 data usually showed noise-like fluctuations around a stationary level, whereas the fluctuations at the Tlamacas station showed a kind of «alive» character, with radon levels that can change rapidly through the day.

In analyzing the radon concentration variations, we focused on the days with moderate eruptive activity, with the data of the eruptions obtained from the CENAPRED webpage. In the period analyzed, moderate eruptive activity was detected for a total of 27 days (39 eruptions, several with rock ejection, some days with 2-4 eruptions per day), 23 of which (32 eruptions) were covered by observations for at least at one volcano station. For 9 days (13 eruptions), we detected a clear decrease (depression) in the radon emission before and during the eruptions (Figure 2, red bars and arrows). The radon release almost ceased for 4 days before the strongest event, that of January 28, 2008 (see Figure 3). During the day of this strongest event, intensive eruptive activity with rock and sporadic lava ejection were detected. Indeed, the period from January 28, 2008, to February 21, 2008, was when the most active eruptive activity was detected. At the same time, before the eruption on January 28, 2008, during the period of January 17-22, 2008, the radon concentrations fell from 900 Bq/m^3 to 200 Bq/m^3 . Furthermore, another total radon decrease (which lasted about 20 h) was detected before an eruption on February 11, 2008. For the following 19 days, which showed moderate eruptive activity, we detected 7 periods of radon decrease, 4 of

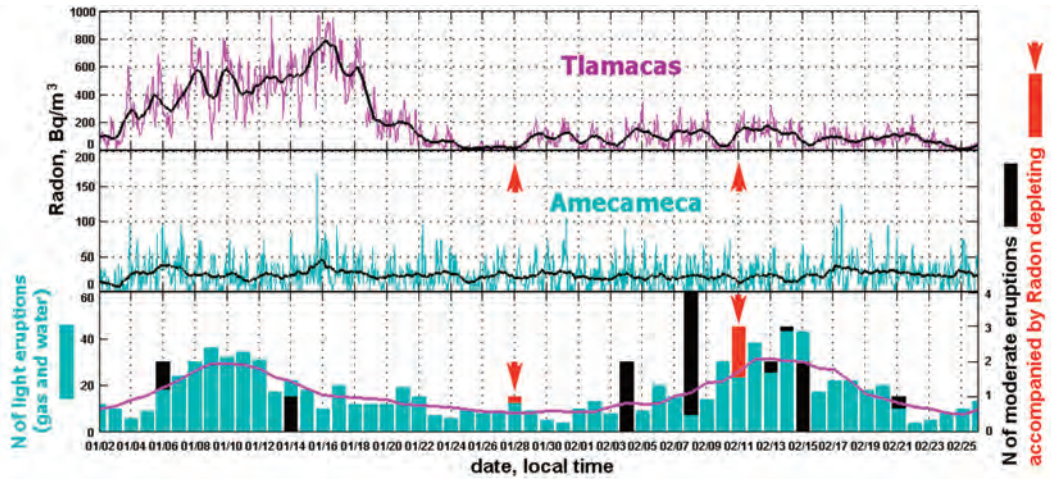


Figure 3. Radon emission monitoring. Top: Tlamacas volcano site. Middle: Amecameca referent site. Monitoring from January to February 2008. Bottom panel: Volcano eruptions. Blue bars, eruptions of gas and water; black bars, moderate eruptions; red bars and arrows, moderate eruptions where radon decreases were observed.

which were seen simultaneously for 2 or 3 of the volcano stations (in all other cases, the data were lost in 1 or 2 stations due to instrumental failure). Again, the most pronounced radon depletion was detected at the Tlamacas observation site, although the radon levels never decreased to zero. In many cases, moderate eruptions were not accompanied by decreases in the radon levels, and no eruptions occurred during some of the other radon decreases. Furthermore, there were better correlations between the radon changes observed at the Paso de Cortes and Tlamacas 2 sites than between these two sites and Tlamacas; the variations in the radon concentrations at the Tlamacas site were sometimes of a very peculiar character.

Summarizing this part of our studies, a couple of phenomena are worth considering. First of all, the radon behavior can be anticipated during moderate eruptions. Unlike the well-known intensifications of radon release before earthquakes, volcano systems are more ambiguous: many studies have reported increased radon emission before major events like explosions and pyroclastic flow, while others have also reported numerous decreases during moderate eruptive activity. Being a heavy gas, radon requires a transport mechanism from the depth where it is generated. This transport is provided by another volcano gas, carbon dioxide (CO_2). Streil et al. [2008] explained the depletion of radon as a shortage of CO_2 due to geodynamic processes in the volcano system. Other, relatively unexpected results were the considerable difference in the radon de-gassing levels, and the periodic absence of correlation between Tlamacas and the other observation sites. Radon emissions depend on a variety of different factors, such as deposits of radioactive elements, presence of active tectonic structures, increased fracturing, soil porosity and penetrability, and meteorological conditions (most importantly, atmospheric pressure). The meteorological conditions at the volcanic observation sites were the same for all of the instruments,

although no information was available about the radioactive deposits in these areas. For the soil composition and penetrability, the Tlamacas site is situated on a solid rock basement, while the Tlamacas 2 and Paso de Cortes sites lie on mellow soil. Thus, the natural activity of radon at these sites was expected to be higher, contrary to the results, as if Tlamacas Mountain was not an active geological structure.

3. Radon survey and gamma-ray spectrometry in the Tlamacas area

A combined study using a radon survey and gamma-ray spectrometry was performed for Tlamacas Mountain and in the nearby areas, to check our hypothesis relating to the anomalous character of the geodynamical processes of Tlamacas Mountain. Gamma-ray spectrometry was chosen as a credible tool to reveal possible radioactive deposits in the area, and to make a decision as to whether the radon emissions were of superficial (i.e. from uranium and thorium deposits) or subterranean (diffusion) origin.

3.1. Equipment and methods

We examined the area by measuring the variations in the total radiometric response, and K, U and Th concentrations using a portable GRS 500 gamma-ray scintillometer spectrometer (Scintrex Ltd.). Each sample site had a series of 12 readings, with a duration of 10 s per reading (0.1 cps), so the total time of observation for every site was 2 min. This technique provides a rational minimization of the signal attenuation and a good signal-to-noise ratio.

The radon survey was carried out using the above-mentioned radon scout instruments. As has been shown, radon emissions in volcano areas can change drastically, even over a single day. To avoid this problem, we limited the duration of our survey to daylight hours on one day, and used

5 identical detectors to carry out simultaneous independent measurements. The test experiment was performed at the Tlamacas site to compare the instrument responses and determine the time domains (sampling) for any independent measurements. We placed the 5 instruments in a hole of about 1 m², at 80 cm in depth, near the Tlamacas station, for 2 days

of correlated measurements, with a 30 min sampling rate. The comparison of the characteristics obtained for the 5 detectors showed the need for correction of one detector (#091), whereas the measurements of the other 4 detectors were comparable and within the instrumental error (Figure 4). The correction coefficient for detector #091 was calculated as

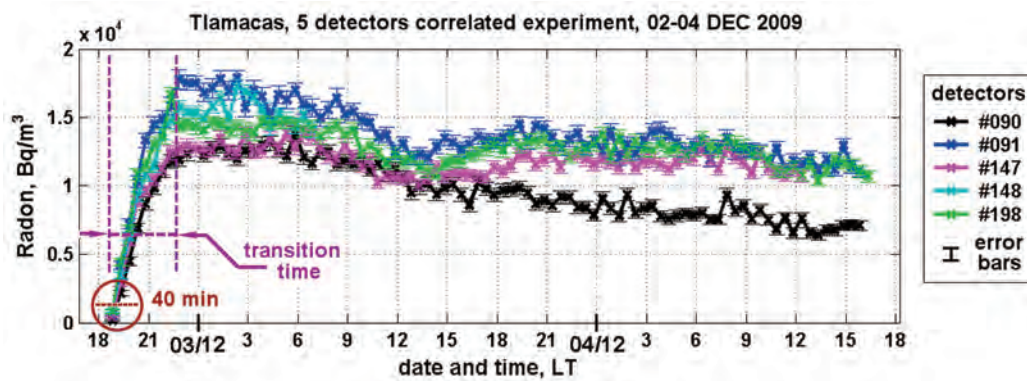


Figure 4. Correlated radon emission measurements for the five detectors tested. Tlamacas station site, December 2-4, 2009 (30 min sampling time).

Reading Point	Easting (km)	Northing (km)	Gamma ray total energy (cps)	Potassium (%)	Equivalent uranium (ppm)	Equivalent thorium (ppm)
(TL1) TL-BL	537.146	2110.417	159.712	0.250	2.066	6.713
TL2	539.091	2108.297	151.332	0.130	2.163	7.672
TL3	539.012	2108.286	196.544	0.054	3.933	8.333
TL4	539.169	2108.339	164.148	0.292	3.844	5.926
TL5	539.135	2108.181	212.951	0.095	5.200	7.407
TL6	538.960	2107.984	163.106	0.021	1.097	5.370
TL7	538.826	2107.817	157.049	0.123	0.954	6.173
TL8	538.663	2107.642	147.066	0.109	0.169	7.593
TL9	538.404	2107.462	167.172	0.024	6.124	8.519
TL10	538.171	2107.477	167.001	0.148	2.968	4.815
TL11	538.025	2107.383	180.070	0.027	2.488	6.019
TL12	537.874	2107.578	171.968	0.180	3.237	10.494
TL13	537.861	2107.794	143.492	0.817	0.680	0.200
TL14	537.733	2108.009	153.104	0.216	2.786	7.222
TL15	537.613	2108.297	157.569	0.125	1.884	7.819
TL16	537.873	2108.684	143.575	0.135	1.417	10.741
TL17	537.434	2108.569	166.170	0.438	0.889	5.787
TL18	537.283	2108.820	164.408	0.161	0.453	7.222
TL19	537.114	2109.041	165.309	0.460	7.026	7.778
TL20	536.870	2109.254	162.966	0.675	4.631	8.148
TL21	536.775	2109.404	162.445	0.398	2.040	7.037
TL22	536.794	2109.631	166.571	0.273	0.983	3.241
TL23	536.913	2109.870	161.624	0.121	0.165	7.639
TL24	536.988	2110.034	157.249	0.191	0.675	7.222
TL25	537.112	2110.222	166.682	0.442	0.822	4.861
TL26	536.561	2110.102	152.483	0.164	1.200	2.778

Table 1. Gamma-ray analysis results (total counts above 0.08 MeV).

TL-BL, Local base station: Paso de Cortes. Easting, Northing, DATUM WGS84, UTM Zone 14.

Point N°	Sensor N°	Latitude (°N)	Longitude (°W)	Radon (Bq/m ³)	Instrument error (%)
1	all (1st test)	19.0856	98.6536	6	98
2	091	19.0832	98.6483	109	38
3	148	19.0808	98.6494	235	24
4	198	19.0676	98.6506	106	35
5	090	19.0761	98.6497	31	71
6	147	19.0742	98.6478	67	45
7	091	19.0725	98.6461	156	33
8	148	19.0703	98.6444	332	20
9	198	19.0706	98.6414	119	33
10	090	19.0697	98.6404	141	33
11	147	19.0675	98.6422	17	100
12	091	19.0650	98.6414	109	38
13	148	19.0618	98.6401	125	33
14	198	19.0592	98.6389	93	38
15	090	19.0597	98.6368	78	45
16	147	19.0594	98.6356	26	71
17	091	19.0606	98.6332	529	17
18	148	19.0628	98.6311	222	25
19	198	19.0650	98.6292	119	33
20	090	19.0664	98.6278	703	15
21	147	19.0675	98.6267	1058	11
22	091	19.0679	98.6281	1244	11
23	148	19.0675	98.6275	2407	8
24	198	19.0675	98.6286	904	12
25	147	19.0669	98.6281	1757	9
26	all	19.0671	98.6282	546	9
27	all	19.0675	98.6278	2789	8

Table 2. Radon concentration results (40 min sampling time).

1.37. An optimal sampling time of 40 min was used, to cover the selected area of our survey from Paso de Cortes to Tlamanca, with a spatial resolution of 200 m to 400 m found between independent points during the daylight hours of a single day.

3.2. Results and discussion

The results obtained are presented in Tables 1 and 2, and shown in Figure 5. Above all, the gamma-ray spectroscopy revealed a moderately enhanced gamma -ray background (Figure 5a) at the top of Tlamanca Mountain, while the levels of uranium and thorium concentrations (Figure 5c, d) were found to be comparable with the average levels (Table 1). The radon survey confirmed the existence of a strong radon anomaly in the area of Tlamanca Mountain (Figure 5f): the radon released at the sites of Tlamanca Mountain was 10-20-fold greater than the background level (Table 2). The lack of coincidence between the maximum of Rn and U or Th, respectively, indicated the subterranean (diffusion) origin of

the radon. The location of the other three maxima of radon concentrations coincided well with three Th/U ratio maxima. This might indicate a proportionality between the ²²⁰Rn and ²²²Rn isotopes in the total radon content, and that their origin most likely lies in superficial deposits of these mentioned elements. Thus, we have demonstrated our assumption relating to the active geological structure in the Tlamanca Mountain area. These results can explain the differences in radon behavior between Tlamanca and the other observational sites, as mentioned in Section 2.

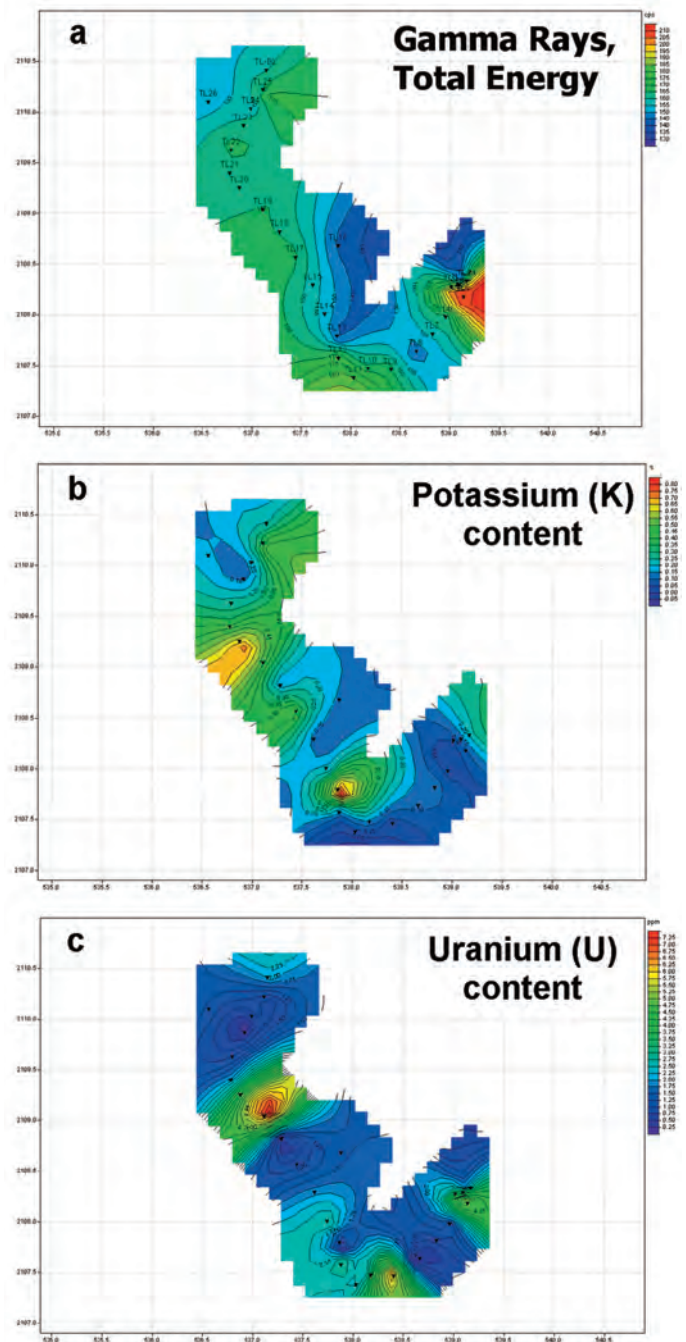


Figure 5a-c. (a) Gamma ray energy (total count above 0.08 MeV); (b) potassium (K; %) content; (c) Uranium (U; ppm) content

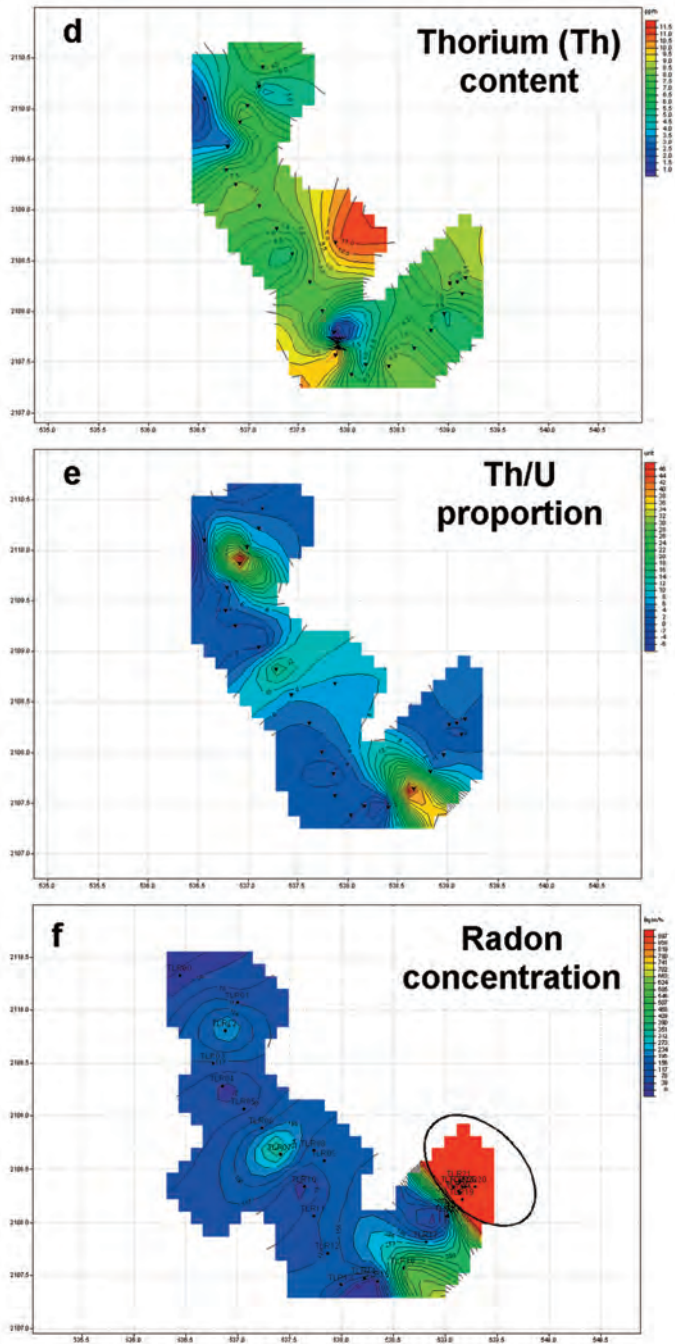


Figure 5d-f. (d) Thorium (Th; ppm) content; (e) Th/U ratio; and (f) Radon concentrations between the Tlamacas and Paso de Cortes stations (top part of the map) and in the area of Tlamacas Mountain (ellipse).

3.3. Limitations of the radon survey method

We should mention some clear limitation of our study. The SHARAD scout detectors were not equipped with pumps: the air sampling was carried out by natural diffusion. Due to this, the effective measurements of the actual values are obtained with some delay, which depends, first of all, on the volume of the pit and the correct radon concentration at the measurement site. Specifically, in our test experiment, the effective transition time was about 4 h (Figure 4). Under similar conditions, the values of radon measured in the first 40 min

(Figure 4, red circle) will be 6-9-fold lower than the expected actual values. This means that our method is eligible in related units, i.e. it can serve to locate sites with anomalously increased radon values, but the absolute radon concentrations will be 6-9-fold greater than the detected values. In this regard, the radon map obtained (Figure 5f) is valid only for comparisons.

Another disadvantage of our method is that the daily values obtained can be affected by the diurnal variation. However, there is no effective way to avoid this drawback. In some cases, a base station can be established in the vicinity of the mapping area, to measure the diurnal variation, which can then be applied for the correction of the radon levels measured at different times. In our case, this method mentioned before is not reliable: all of the base stations should be established at least several days in advance of the survey, and remain at the site to obtain enough data for solid statistics. Furthermore, in «aggressive» environments such as volcanoes, diurnal variations can get lost among high levels of noise-like changes (which was also in our case).

4. Radon and atmospheric electricity: new aspects of lithosphere-atmosphere coupling?

4.1. New view on the volcano and seismogenic geomagnetic emissions

The intensive radon release for Tlamacas Mountain encouraged us to revise our earlier discovered phenomena [Kotsarenko et al. 2007, 2008]: long-term noise was observed in the geomagnetic field measured at the Tlamacas station (Figure 6). To estimate the noise contribution, we compared the noise character for the Tlamacas station registered during a highly perturbed day with a referent signal contaminated by electric welding measured 200 m away from the welding source in Campus Juriquilla, Querétaro, during the construction of a new building. The natural perturbations observed for Tlamacas were equal or even higher in amplitude compared with the intense corona discharge (electric welding currents of up to 100 A).

How can this be connected to the observed radon emission phenomena? Increased radon release produces high ionization in the atmosphere [Pulinets and Boyarchuk 2004], which changes the electric field profile. Moreover, in the case of the Tlamacas station, an equivalent electric circuit between the Earth and thunderstorm clouds (8-12 km) changes strongly and gets much shorter for the altitude of Tlamacas Mountain (4 km). Thus, any heterogeneity in the ground surface becomes the source of micro-discharges, which are detected as integrated electromagnetic noise by the magnetometer.

4.2. Thunderstorm lighting as a possible indicator of forthcoming earthquakes

Another phenomenon of the same nature is often reported by witnesses: the frequency of lightning in the

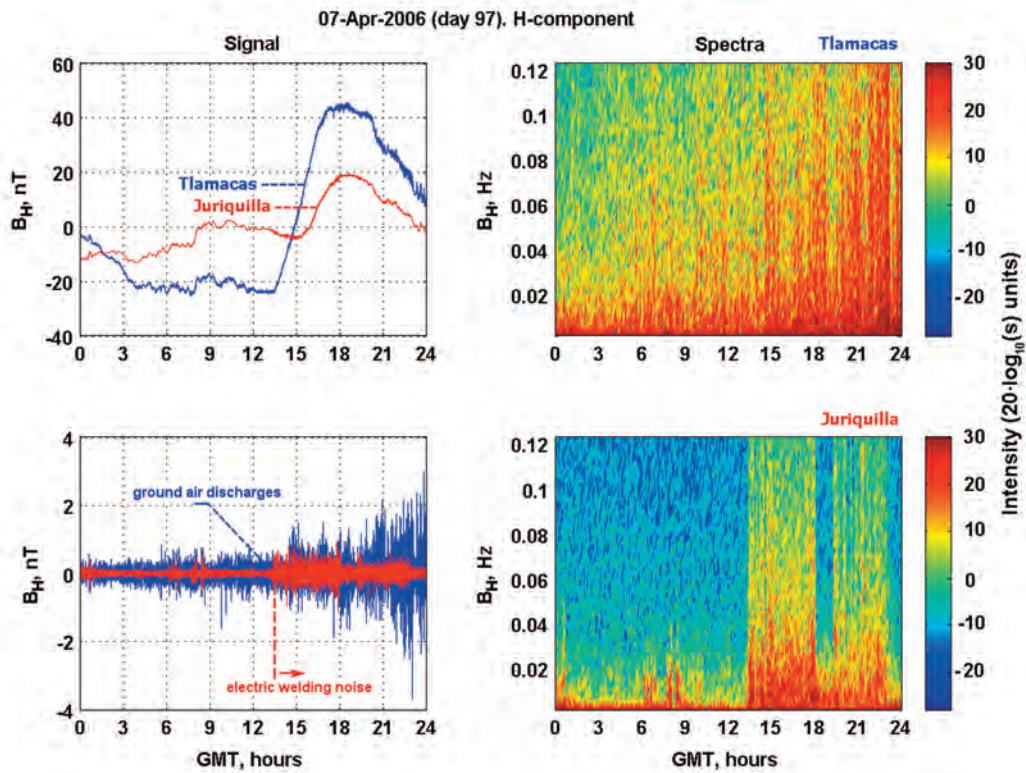


Figure 6. Geomagnetic signals. Left, top, Original signals; Left, bottom, Filtered signals. Blue, Tlamacas station; red, referent Juriquilla station. Right: Spectra measured at Tlamacas (top) and at the referent Juriquilla station (bottom, red).

Tlamacas Mountain is higher than in other sites (one such lightning strike destroyed our magnetometer and burned out a CPU despite the presence of a lightning-rod on the station building). This can also be explained in terms of the atmosphere electricity: the increased ionization and closeness to the thunderclouds attracts lightning, as happens during thunderstorms. These phenomena have significant scientific applications.

First of all, Tlamacas Mountain becomes a unique natural laboratory for experiments with atmosphere electricity. In terms of lithosphere-atmosphere coupling, Tlamacas Mountain performs as a natural reactor of ionization that modifies its electrical profile, provoking intensive integrated noise-like currents (by means of micro-discharges) in the near-surface air layer, and enhanced thunderstorm activity between the Earth surface and the thunderstorm clouds.

Thus, the presented concept can re-interpret the noise-like geomagnetic emissions observed before major earthquakes in the earthquake preparation zone. The origin of these mentioned emissions might be attributed to the proposed mechanism of micro-discharges in the highly ionized superficial air.

Finally, we can propose the following hypothesis: monitoring of lightning activity in seismo-active areas can be of help for the forecasting of major earthquakes. The preliminary confirmation of this statement came from a

study dedicated to the Alum Rock earthquake (October 30, 2007, California, USA) [Bleier et al. 2009]. This study examined the lightning activity in the earthquake preparation area, to remove the interference of thunderstorm-related pulses from geomagnetic signals. The maximum number of lightning strikes was detected the day before the earthquake in the area of the Sierra Nevada mountains. We believe that this is a similar example in nature to that presented in our study.

5. Conclusions

Our results of the radon monitoring at three different sites of Popocatepetl Volcano mountain and a referent site revealed that radon depletion preceded nine cases of moderate eruptive activity among 23 total events. The most pronounced reaction was observed at the Tlamacas observational site. The average radon concentration at the Paso de Cortes and Tlamacas two sites was significantly lower in comparison with that at Tlamacas; the radon variation in these other two sites has many specific features, while for the Tlamacas site, the radon behavior shows a more individual character.

The combined study using the radon survey and gamma-ray spectrometry revealed an anomalously increased diffusion radon emission localized in the area of Tlamacas.

A new concept is proposed regarding lithosphere-atmosphere coupling in the case of Tlamacas, which appears

to be similar in nature to a shortened electrical circuit from the Earth to the thunderstorm clouds (high-altitude mountains), such that the enhanced ionization caused by intensive radon release can explain, in a novel way, the noise-like geomagnetic emissions observed before destructive earthquakes. We also propose the study of thunderstorm activity in the earthquake preparation zone as a promising tool for the forecasting of destructive earthquakes.

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