Possible Structural Connection between Chichón Volcano and the Sulfur- Rich Springs of Villa Luz Cave (a.k.a. Cueva de las Sardinas), Southern Mexico

Laura Rosales Lagarde[1], Penelope J. Boston[1, 2], Andrew Campbell[1], and Kevin W. Stafford[1]

[1] New Mexico Institute of Mining and Technology, 801 Leroy Place 2421, Socorro, New Mexico 87801 USA, lagarde@nmt.edu
[2] National Cave and Karst Research Institute, 1400 University Drive, Carlsbad, New Mexico 88220 USA

Abstract

Regional strike-slip faults may serve as groundwater flow-paths from the active Chichón Volcano to the Villa Luz Cave (a.k.a. Cueva de Las Sardinas, CLS). In this cave, located near Tapijulapa, Tabasco, several springs carry hydrogen sulfide. Previous studies have linked the CLS spring sulfur source to basinal water and an alkaline active magma volcano, but the groundwater flow paths still need to be reviewed. Understanding the sulfur origin in the cave will provide insights into the possible sources, the extreme microbial environment, the sulfuric acid speleogenetic mechanism (i.e. creation of caves by strong acid dissolution), the subsurface water-rock interactions and groundwater flow paths in the area. The volcano and CLS location in the Chiapas Strike-slip Fault Province, suggests a left-strike slip fault may be serving as a groundwater flow path, allowing deep-source magmatic water to contribute sulfur to the water that is dissolving the limestone at CLS. Detailed geological mapping of the surface and the caves in-between, coupled with chemical analyses of the cave and spring waters may help to prove this connection.

Introduction

Although Villa Luz Cave (a.k.a. Cueva de Las Sardinas, CLS) is forming in limestone, its groundwater flow-paths may be connected to the active Chichón Volcano by regional, siniestral strike-slip faults. Although previous studies suggest a possible contribution of volcanic sulfur to the cave waters [Hose *et al*., 2000; Spilde *et al*., 2004], the groundwater flow-path to accomplish this was not suggested. Although CLS is 50 kilometers east from the active Chichón Volcano, lateral faults in the area and the structures associated with it, can provide the necessary flow path for the sulfur-rich water. The study of other sulfur springs between the cave and the volcano will also help to provide evidence to support this hypothesis. Specifically the location, the geology, the water chemistry and isotope composition of sulfur, oxygen and hydrogen at three sulfur spring areas will be acquired and analyzed in order to accomplish this goal.

Chichón Volcano produced an unusually sulfur-rich magma in its last explosive eruption in 1982, leaving an active hydrothermal system. The unusually high sulfur concentration of that eruption has not yet been explained. Nevertheless, evaporitic subsurface deposits may influence Chichón hydrothermal water composition and/or act as a sulfur source to the Las Sardinas Cave sulfur springs. This cave is typified by the high sulfur concentration of most of the springs present in the cave. These conditions produce a sulfur-rich microbial environment resembling deep-sea hydrothermal-vents [Boston *et al*., 2006]. The hydrogen sulfide present in the cave reacts with the limestone enlarging the cave by the sulfuric acid speleogenetic mechanism (i.e. creation of caves by strong acid dissolution). The study of this system will provide insight to this process.

The understanding of the sulfur origin to Villa Luz Cave and sulfur springs in the area will help to identify the relevance of the possible sources as well as the subsurface water-rock interactions occurring.

A review of the geological setting and the main characteristics of the Chichón Volcano and the Villa Luz Cave, followed by the proposed methodology to test the volcano-cave groundwater connection will be presented in this paper. Further results and conclusions are not yet available because the main part of this project is still in progress.

Geologic Setting

Location: Villa Luz Cave is located 50 km east of Chichón Volcano, near the border of the states of Tabasco and Chiapas, southern Mexico (Figure 1). In addition to Chichón, Villa Luz Cave sulfur-rich spring water can be influenced by the Chiapas-Tabasco Oil and Gas Fields with high-sulfur content to the north, the ~5 Ma Santa Fe and Victoria granodiorite intrusive rocks to the west, and older andesitic flows to the north and southwest of the area.

Structural setting: CV and CLS are located in the north of the Strike-slip Fault Province defined by Meneses-Rocha, [2001]. The Strike-slip Fault Province occupies the Sierra de Chiapas, to the north with elevations ranging from 100 to 2000 m.a.s.l. This province is formed by upthrown and downthrown blocks, formed during a transtensional phase, bounded by lateral strike-slip faults. Northwest trending en-echelon anticlines with middle Cretaceous and Paleogene rocks in their center are present in most of the upthrown blocks while tectonic basins filled with Cenozoic rocks are present in the downthrown blocks [Meneses-Rocha, 2001]. The aforementioned author states that syndepostional tectonism is evidenced by local unconformities, thickness changes and lithologic variations along structural trends. The orientation of faults in this province is the basis for a further subdivision [Meneses-Rocha, 2001]: a) a western area, with variably oriented faults; b) a central area, with northwest oriented faults and, c) an eastern area, with west oriented faults. The eastern most part of this province is where our study area is located (Figure 2). The detachment surface of the central and western areas is comprised of Callovian salt deposits, while in the eastern area a Lower Cretaceous anhydrite (Cobán

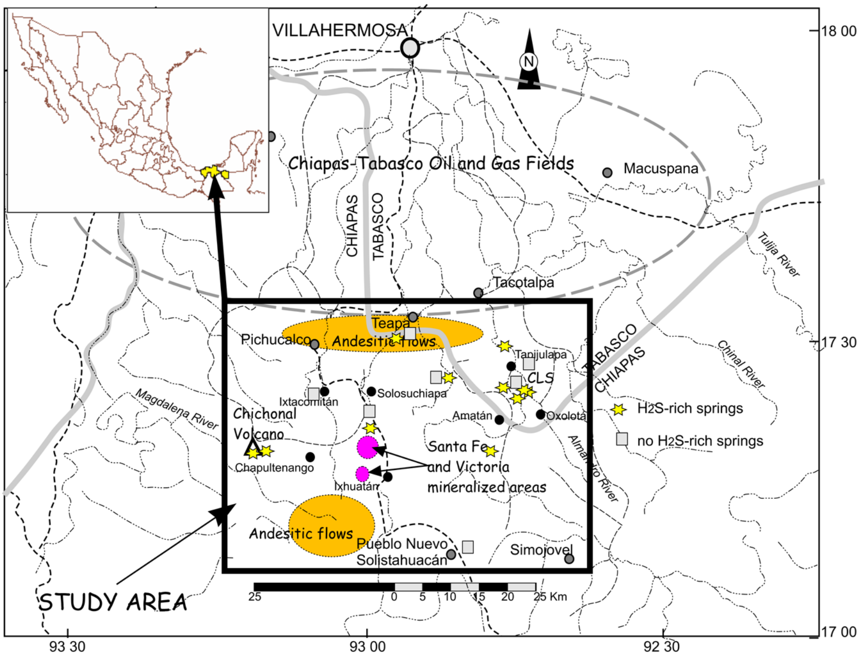

Figure 1. Location of the CLS (Cueva Las Sardinas or Villa Luz Cave) and Chichón Volcano (CV). H_2S-rich springs are presented by stars while no-H_2S springs are presented by squares. The possible sulfur sources to the sulfur-rich springs in CLS are also shown.

Formation) detachment is also identified (besides the aforementioned detachment level) (Figure 3). These detachment levels could provide sulfur to the groundwater feeding Chichón Volcano 1982 magma and/or the Villa Luz Cave springs. A basement involvement in some of the faulting is evidenced by the presence of Pliocene-intrusives (Santa Fe granodiorite, 5 Ma) and Pliocene-Quaternary volcanoes (Chichón Volcano) at the ends of some faults [Capaul, 1987; García-Palomo *et al.*, 2004; Meneses-Rocha, 2001].

Geologic history: Rocks from Cretaceous to Quaternary age outcrop in the area (Figure 4 and Figure 5). The basement is considered to be Paleozoic granites that crop out in the Chiapas massif and metamorphosed sediments south and east from the area, respectively. Paleozoic granitoids and Mississippian to Permian slightly metamorphosed sediments (shale, sandstone and limestone) are also present. The post-Permian - Upper Jurassic opening of the Gulf of Mexico produced discordant conglomerate, sandstone and shale-filled half-graben structures, probably syntectonic to salt and evaporitic deposits. Evaporite deposit extension in the area, shown in Figure 3, is responsible for the distribution of compressional salt tectonics [García-Molina, 1994] and for the deformational response in the different structural provinces. Basinal to shallow platform carbonates, to littoral and alluvial fan environment sediments interfingered during upper Jurassic times. The basinal facies served later as a hydrocarbon source. Carbonate sediments dominated Cretaceous deposition from the Yucatan Platform

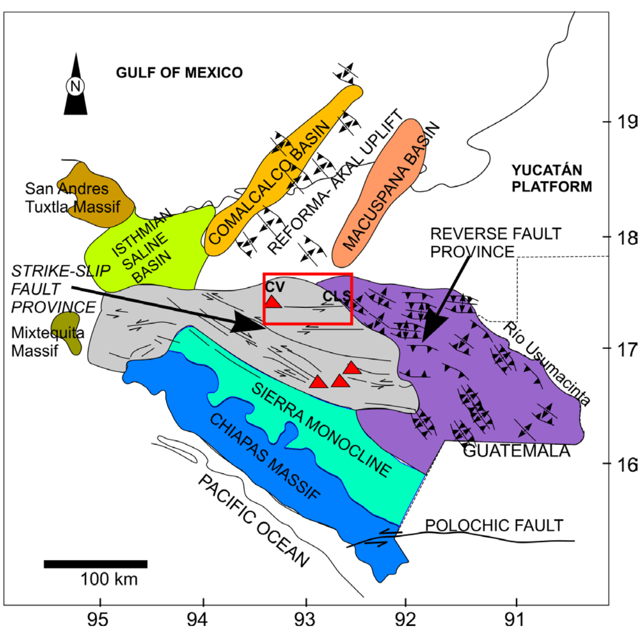

Figure 2. Structural provinces present in the study area. Chichón Volcano (CV) and Villa Luz Cave (CLS) are located in the Strike-Slip Faults Province. Volcanos from the Chiapas Volcanic Belt or Arc in the area are shown as triangles (Modified from Meneses-Rocha [2001]).

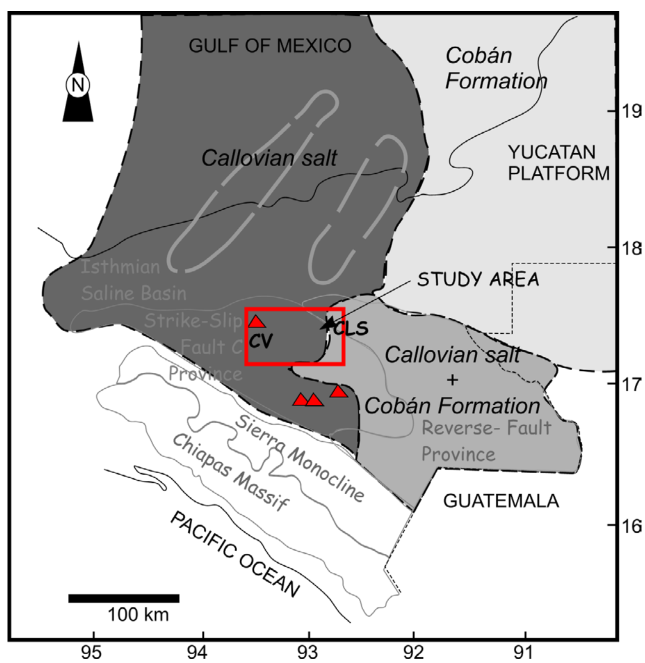

Figure 3. Type of evaporite deposit underlying the area (Callovian salt, halite or Cobán Formation, anhydrite). These deposits were one of the major controls on defining the structures present (modified from Meneses-Rocha [2001]).

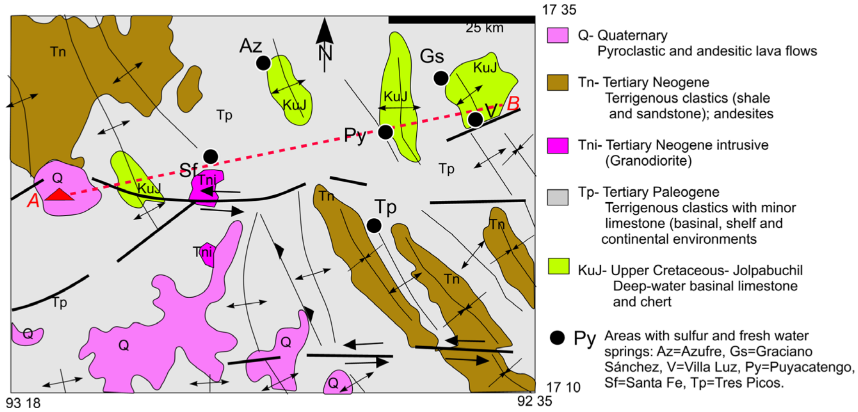

Figure 4. Generalized geologic map of the study area, showing the location of Chichón Volcano (CV) and Villa Luz Cave (CLS), as well as other sulfur spring areas. The orientation of the geologic section shown in Figure 5 is also shown (Modified from INEGI [1983] and Meneses-Rocha [2001]).

to the west of the Chiapas range, unconformably covering older rocks. This age sedimentary environments vary from supratidal to reef and pelagic. Between Paleocene and middle Eocene, during the Laramide orogeny, the area was subject to gentle deformation causing terrigenous sediments and interfingered carbonates to disconformably deposit in flexural basins [Meneses-Rocha, 2001]. The Cayman Trough insertion and Polochic-Motagua Fault began at the end of the Paleocene forming normal and lateral faults. From Late Eocene to Early Miocene, the Strike-slip Fault province movement along the faults was predominantly vertical, changing to sinistrally transcurrent at the beginning of the Middle Miocene (transtensional phase). During the late Miocene-early Pliocene, a coarse-continental sequence was deposited in response to normal block faulting of the basement caused by the shift of the main bounding faults. Meanwhile carbonate platform units deposited on the Yucatan platform and some parts of Chiapas. At the end of the Pliocene, a transpressive episode deformed some of the previously formed basins. This event was related to the rise of the Neogene Chiapas fold and thrust belt by basal decollement movement over the Jurassic salt, and recession of the shoreline to its present position. This last compression event relates to the intrusion of granitoid bodies. During the Quaternary, volcanic sediments were deposited in angular unconformity on the continental sediments.

The total sinistral shear across the Strike-strip Fault province is estimated to be of approximately 70 km, and the individual faults in this province has a displacement greater than 16 km [Meneses-Rocha, 2001]. The importance and participation of the structures present in the groundwater control are not fully understood yet.

Volcanic rocks associated with an arc have been present from the Permian until present [García-Molina, 1994].

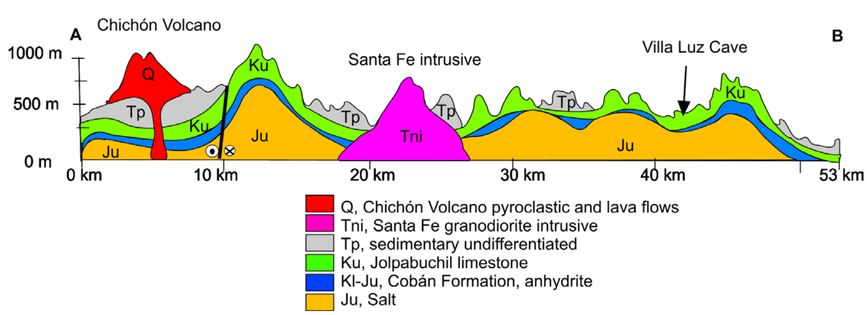

Figure 5. Geologic section of the study area showing a simplified interpretation of the geology (Figure 4, vertical thickness of the formations are not in scale). Ju=Upper Jurassic, Kl=Lower Cretaceous, Ku=Upper Cretaceous, Tp=Tertiary Paleogene, Tni=Tertiary Neogene intrusive, Q=Quaternary.

Chichón Volcano

Chichón or Chichonal Volcano is the youngest and western most K-rich andesitic volcano of the Chiapas Volcanic Belt or Arc (Figure 1 and 2), [Macías *et al*., 1997], with deposits at least 8000 years old [Espíndola *et al*., 2000]. Located in a still-debated tectonic setting [Espíndola *et al*., 2000; De Ignacio *et al*., 2003], it is proposed as one of the possible sources for the CLS cave sulfur-rich water springs [Hose *et al*., 2000; Spilde *et al*., 2004]. The Chichon volcanic cone was built on folded Cretaceous dolomitized limestone underlain by Jurassic evaporites and covered by alternating sequences of Tertiary shale and marl [Macías *et al*., 1997], (Figure 5). Structurally, this volcano is located in a strike-slip regime, at the junction of three main structures (Figure 6): (1) the Chapultenango extension Fault System; (2) the NW-SE trend Buena Vista Syncline; and (3) the San Juan Fault System (strike-slip), with an E-W orientation. The latter is proposed as the K-alkaline magma feeding-system [Macías *et al*., 1997; García-Palomo *et al*., 2004]. These structural features control the pattern of rivers and determine the topographic irregularities around the cone [Scolamacchia and Macías, 2005].

After its last eruption, in March-April 1982, a crater lake formed and the associated hydrothermal system was redefined [Taran *et al*., 1998; Rouwet *et al*., 2004] with active fumaroles depositing elemental sulfur (Figure 7). Luhr and Logan [2002] estimate that 2.2 x10^{13} g of S were emitted on the 1982 CV eruption, from which 58 wt.% of the sulfur was present as anhydrite prior to eruption, with the remainder in a vapor phase, with H_2S/SO_2 ≈9. These authors also discard a sedimentary provenance to the anhydrite based on sulfur isotopes, supported by chemical evidence indicating absence of hydrothermal fluid interaction with the underlying evaporites or basement rocks [Taran *et al*., 1998]. Nevertheless, Espíndola *et al*., [2000] suggest that the high-sulfur magma of the 1982 eruption, and probably previous eruptions, was created by a mafic magma injection into the underlying limestone.

Although the 1982 eruption produced anhydrite-rich pyroclastic deposits [Luhr and Logan, 2002; Taran *et al*., 1998], the hydrothermal system until 1997 showed

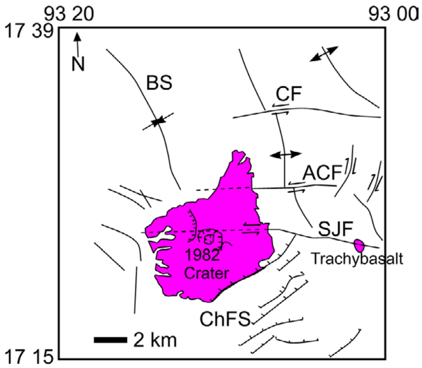

Figure 6. Plan view of Chichón Volcano (CV), showing the E-W lateral San Juan Fault (SJF) interpreted to control the magma-feeding system (modified from García-Palomo *et al*. [2004]) and which may serve as a groundwater flow-path for sulfur water from CV to Villa Luz Cave. Other major structures are: CF=Caimba Fault, ACF=Arroyo de Cal Fault, ChFS= Chapultenango extension Fault System, and BS=Buenavista Syncline.

relatively low sulfur content. Between 1998 and 1999, sulfate concentration increased in the lake water, decreasing in 2000 while H_2S/SO_2 ratio increased in the fumaroles [Rouwet, 2004; Taran *et al*., 1998; Tassi *et al*., 2003]. The variability of the sulfur concentration in the hydrothermal system may reflect magma movement [Horwell *et al*., 2004; Taran *et al*., 1998].

Villa Luz Cave (a.k.a. Cueva de las Sardinas, CLS)

CLS is located on the northeast side of the study area (Figure 1). The cave formed on a folded block of Cretaceous micritic limestone bounded to the south by a normal fault, with structure probably controlling the cave inlet's location [Hose *et al*., 2000]. Due to the normal fault orientation, it may represent a permeable conduit connecting the cave to the San Juan lateral Fault at the Chichón Volcano (Figure 4).

The first studies of CLS focus mainly on the fish present [Gordon and Rosen, 1962]. Pisarowicz [1994] attracted international attention to the cave, resulting in further studies [Estrada B. and Mejía-Recamier, 2005; Hose *et al.,* 2000; Langecker *et al*., 1996; Northup *et al*., 2002; Plath and Heubel, 2005; Plath *et al*., 2006; Spilde et al., 2004; Boston *et al*., 2006]. Plath *et al*., [2006] also present a brief review of the studies history at Cueva de las Sardinas and a summary of the cave fish research. Hose and Pisarowicz, [1999] provide a detailed map and description of CLS, (Figure 8) while Hose *et al*., [2000] comprehensively describe CLS, including the cave's speleogenetic mechanism, based on detailed morphologic and chemical measurements. They also conducted preliminary biological analyses emphasizing the microbiological importance in the cave development. At least 26 springs have been identified in CLS (Figure 8). Based on their chemical nature and physical appearance, Hose *et al*. [2000] classify the springs in the cave as two end members: A and B. End member A is characterized by $[H_2S]$= 300-500mg/l and $[O_2]$< 0.1mg/l. This water is slightly supersaturated with calcite and undersaturated in gypsum and dolomite; recognizable in the cave by elemental sulfur coating the walls above the spring (Figure 9), white bacterial filaments on the wet rock surfaces, and pyrite deposits on the sediments/rocks covered by water. Spring water B has $[H_2S]$ <0.1mg/l and $[O_2]$ <4.3mg/l. These inlets are characterized by travertine precipitation and red-yellow iron oxides, calcite and dolomite supersaturation and undersaturation in gypsum. AB water results from the mixture of the first two springs end members. AB composition water is the most abundant present in the cave (pH, P_{CO2} and SI similar to B and characterized by white coloration probably produced by colloid-size sulfur particles [Hose et al., 2000]. Based on total dissolved solids and general chemistry, a similar origin and composition was proposed for the A and B springs inside the cave, suggesting oxidation of H_2S in the B springs before arriving to the CLS. The causes or controls for the water oxidation are still unknown. In this paper, we will refer mainly to the A-member springs as sulfur-rich springs, focusing on its possible connection to the Chichón Volcano.

In Villa Luz Cave, sulfur-rich springs are actively dissolving bedrock (i.e., Sulfuric Acid Speleogenetic mechanism) while supporting abundant sulfur-based microbial life and providing energy to the cave ecosystem [Hose et al., 2000]. Hydrogen sulfide degassing from the spring water oxidizes to elemental sulfur or sulfuric acid. The latter one reacts with the limestone to produce selenite crystals or gypsum paste (Figure 9).

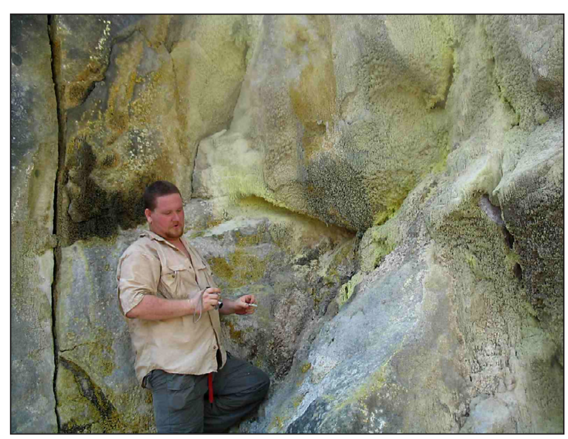

Figure 7. A view of Chichon Volcano crater lake from the west rim and sulfur deposits on the internal west crater wall associated to the fumaroles.

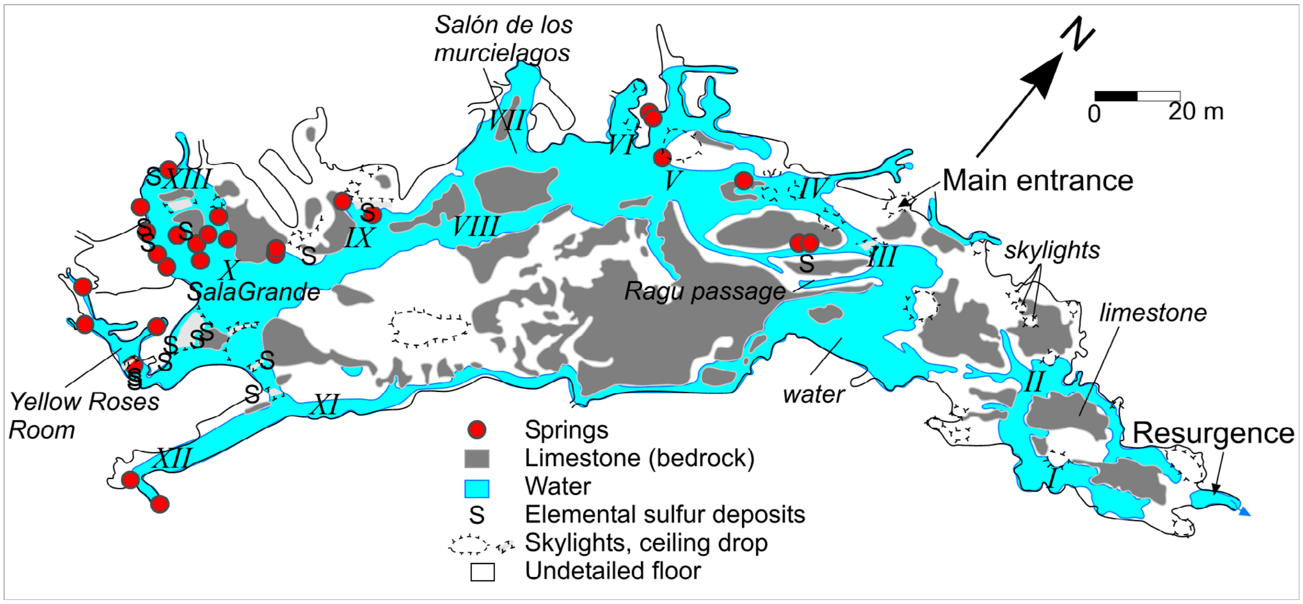

Figure 8. Simplified plain view of Villa Luz Cave (a.k.a. Cueva de las Sardinas, CLS) emphasizing the location of springs (red circles), streams skylights and limestone columns. The position of the main entrance, resurgence and areas with elemental sulfur is also included (Surveyed by Pisarowicz et al, [1998]; map modified with permission of Bob Richards). The approximate location of chambers (*I-XII*) from Gordon and Rosen [1962]; Plath et al. [2006] is integrated for reference.

Although different sources have been suggested for the sulfur origin of the cave springs, the dominant hypothesis is that basinal water [Hose et al., 2000] is influenced by the active, anhydrite-rich magma of Chichón Volcano [Spilde et al., 2004]. Nevertheless, neither the relationship with other possible sulfur sources in the area, nor the groundwater flowpaths, nor the controls on this flow are well understood. Sulfur-rich oil and natural gas fields [García-Molina, 1994], a Tertiary age skarn system (Figure 1) [Castro-Mora, 1999; Pantoja-Alor, 1968], a thick underlying evaporite layer [García-Molina, 1994], and decomposition of organic matter under anoxic conditions [Stoessell et al., 1993] could also be potential hydrogen sulfide producers.

Previous evidence of connection: Based on He isotopic relations of one gas sample and water samples from four springs Spilde *et al*. [2004] determined that at least 22% of the gas at CLS has a magmatic component (mixing of mantle and crustal sources), while 6% of the water has a hydrothermal origin, and the rest of meteoric origin.

Several other sulfur springs have been identified between CV and CLV (Figure 1). From the identified springs, only those at Villa Luz Cave, at Cueva Luna Azufre [Pisarowicz., 2005] and a small cave north from CLS [Siegel and Amidon, 2006] (GS, Graciano Sánchez in Figure 4), have been found to be associated with caves; the rest of them are either covered by alluvial deposits, underwater and/or too small to be humanly entered. The only sulfur-spring that has been further studied, besides the ones at CLS, is at El Azufre, Teapa, Tabasco [Hose, personal communication; Nencetti et al., 2005; Spilde et al.,

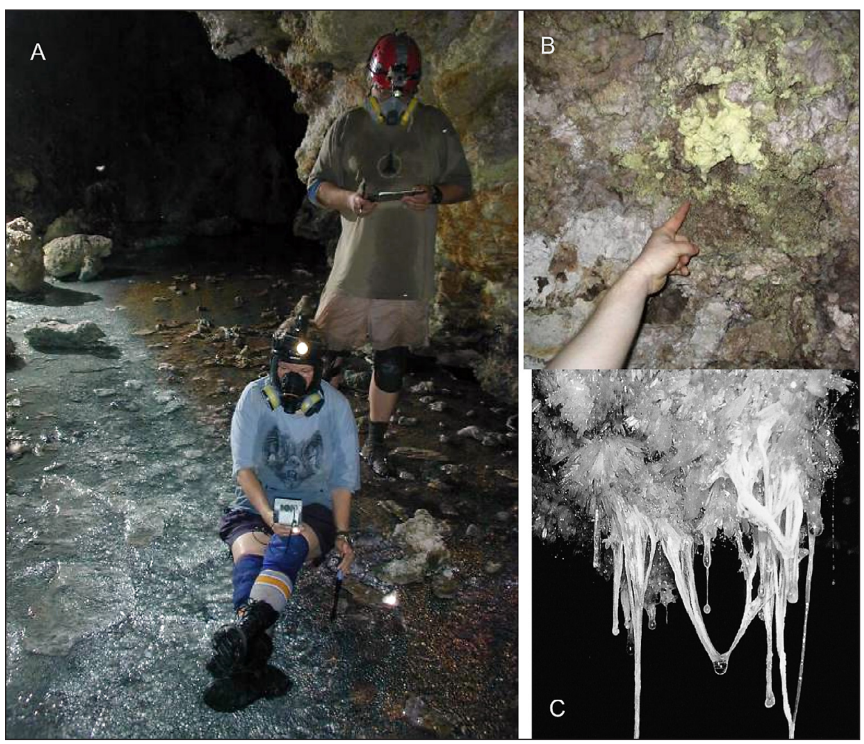

Figure 9. Photographs of Villa Luz Cave: A. End member springs, H_2S-rich in the left and H_2S- poor to the right [Hose *et al*., 2000], (Photograph by Kenneth Ingham); B. Sulfur deposits on the ceiling, associated to H_2S-rich springs; and C. Selenite deposits with biofilms (snottites) (Photograph by Kenneth Ingham).

2004; Taran, personal communication]. Hose [personal communication] found a good correlation in the sulfur concentration and other chemical parameters of El Azufre area sulfur-rich springs with those of Villa Luz Cave (Figure 4). Also, El Azufre springs were the only ones rich in H_2S from those sampled by Nencetti *et al*. [2005]. Based on gas and/or water samples from nine springs in the Sierra de Chiapas, south of the study area, Nencetti *et al*. [2005] proposed a close association between the thermal spring location and the Cenozoic volcanic centers. They also suggested a strong fault and fracture control on the spring presence, as well as a mixture between shallow aquifer water and a more saline member, with higher rock-water interaction.

Therefore, a geologic and geochemical characterization of the springs between Chichón Volcano and Villa Luz Cave will help to determine the permeability/connectivity between both, as well as possible groundwater fluid flow-paths which may be of help to understand water and oil migration in the area.

Methodology

The determination of the Chichón Volcano (CV) - Villa Luz Cave (CLS) connection is part of the first author's Ph.D. studies which the development of this project is still in process. The project general methodology and justification is discussed below.

Background: A review of the geological and water chemistry information available in the area from different sources, including surface and subsurface geology, river water chemistry and weather conditions provide initial data for the project. Subsurface stratigraphic variations will be determined by log correlation of available wells. The characteristics at depth of the structures present in the area will be determined based on the available interpreted seismic sections.

Preliminary field and laboratory analysis allowed the identification of other sulfur springs areas in-between CLS and CV. Based on these data three smaller regions with sulfur springs were selected for further geological mapping and water sampling: 1) Santa Fe region; 2) Puyacatengo region; and 3) Villa Luz region (Figure 4). This will provide an east-west section from the volcano to the cave where concentration variations can be determined, for example sulfate, H_2S, cations concentration, etc.

Geological mapping: Geological mapping will focus on the selected study regions. Since the study area is highly vegetated, the mapping techniques to be used in the selected study regions are outcrop mapping combined with geologic sections focusing on lithologic contacts and structures [García-Palomo *et al*., 2004; Marshak and Mitra, 1988]. Previously identified structures within the study area [Meneses-Rocha, 2001; INEGI, 1983, Castro-Mora, 1999], will also be reviewed in the field. Lineations controlling the surface and groundwater movement will be determined at different scales. Satellite radar images will be analyzed to determine preferential regional lineation direction (Figure 10), while cave maps in the selected regions will be studied to determine preferential local groundwater flow directions. Instances of caves and karst surface terrain will be documented and serve as alternative outcrops in highly vegetated areas of the study area [Dasher, 1984]. Available cave maps and locations from the Caves of Tabasco Project of the National Speleological Society will enable further geomorphologic and structural evaluation. Joints and structures will be measured at an outcrop level close to the springs to determine main structures involved and its relation to major structures.

Rock samples will be taken for petrographic analyses. Samples with sulfides, sulfates or elemental sulfur will be processed for sulfur stable isotopes.

Springs identified will be classified according to Bögli [1980] and major field parameters measured on each of them, including: pH, temperature, conductivity, dissolved oxygen, alkalinity. Air temperature measurements will help to detect the presence of hydrothermal water, discarding altitude differences. The information collected at each spring will include its geographic location, the host lithology, associated geological characteristics and classification.

Diagenesis in some cored-rock samples of oil/exploratory wells in the area will be examined in thin sections to provide the extent of sulfur mineralization/sulfate reduction and/or related processes occurring at depth and their relative timing (samples provided by Exploration and Production Department of the Mexican Oil Company, PEMEX).

Water sampling and chemical analysis: According to the classification of the springs in the selected regions and their major chemical parameters, some of them will be selected for further

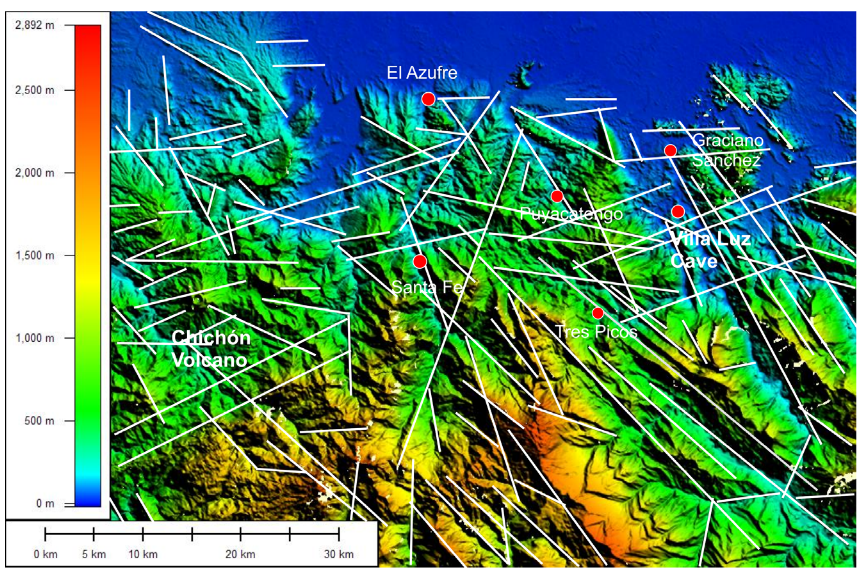

Figure 10. Major lineations on a radar image of the study area (white lines), showing the location of Chichon Volcano, Villa Luz Cave, and other sulfur springs regions (Santa Fe, Puyacatengo, El Azufre, and Graciano Sánchez sites). Darker color represents lower elevation above sea level (Radar image from http://www.dgadv.com/dowdem/, modified with Global Mapper). [The color elevation scale on the version on the CD is easier to understand.]

sampling and water analysis. Rainwater and produced water from producing oil wells in the area will also be analyzed for comparison. The water samples will be analyzed for cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) and anions (SO_4^{2-}, Cl^-, F^-, NO_3^-) [Greenber *et al*., 1992]. These are the most commonly used elements to classify water because their concentration in water reflects water-rock interactions and groundwater sources [Appelo and Postma, 1993]. Cation samples will be acidified with nitric acid and analyzed by ICP-OES. Samples for anions will be filled without air space and analyzed by Ion Chromatography. Separate samples will be collected for δD- δ^{18}O, and total carbon analysis.

Sediment and rock samples of the sulfur springs will be taken for the stable isotopic analysis of the sulfides precipitated, while the dissolved sulfate will be precipitated with barium chloride [Bottcher, 1999; DeCaritat, 2005; Rajchel, 2002]. δ^{34}S of both precipitates and δ^{18}O of the sulfate precipitate will be determined, to compare the source and reactions occurring in the H_2S and the water sulfate, determine the source of oxygen to the sulfate and the biological participation in these reactions [Hoefs, 2004].

Expected results

D-O isotopes of the analyzed water samples will help to determine the input of meteoric (rain) water, and evaporation/condensation in the sulfur springs of Villa Luz Cave and along the east-west transect.

Sulfur isotopes are one of the main tools that will be used to determine the possible connection between the cave and the volcano. Isotopic concentration is expressed as δ^{34}S [Hoefs, 2004] relative to CDT (Canyon Diablo Troilite Standard). Mantle δ^{34}S is near 0‰, so Chichon Volcano sulfur values may be close to this value, unless the magma is assimilating sedimentary anhydrite, while if sulfates are just coming from the subsurface anhydrite, they will show limited variability (δ^{34}S~+17± 2‰) compared with the values spread in marine sulfides (-5 to –35‰) [Condie, 2005]. Since microorganisms strongly prefer the lighter isotope, ^{32}S, sulfate reducing bacteria will produce negative δ^{34}S values in organic sulfides [Canfield, 2001]. Therefore biological participation in the cave or along the groundwater flow-path may be identified. The relation Na-Cl in the groundwater may indicate the influence of the Callovian-salt in the groundwater, which may be related to an increase in permeability along the detachment level or salt ascension.

The coupling of all the elements mentioned above will provide a better description of the relationship/connectivity between the Chichón Volcano and the Villa Luz Cave. General chemistry and stable isotopes analysis of some preliminary samples are being analyzed in order to determine a better sampling/mapping strategy for the main field work planned to start on January 2007.

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