

Chemical evolution of thermal springs at Arenal Volcano, Costa Rica: Effect of volcanic activity, precipitation, seismic activity, and Earth tides

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Abstract

Arenal Volcano in NW Costa Rica, Central America has been active during the last 37 years. However, only relatively low temperature springs have been identified on its slopes with temperatures less than around 60 °C. The springs are clustered on the NE and NW slopes of the volcano, close to contacts between the recent and older volcanic products or at faults that intercept the volcano. This volcano is located in a rain forest region with annual rainfall averaging around 5 m. During the last 15 years, the temperature and chemical composition of 4 hot springs and 2 cold springs have been monitored approximately every 3 months. In addition, two more thermal sites were identified recently and sampled, as well as two boreholes located on a fault NE of the volcano. Scatter plots of chemical species such as Cl and B suggest that the waters in these discharges belong to the same aquifer with a saline end member similar to Río Tabacón at the beginning of the study period (1990) and the deeper borehole (B-2) in 2004. The waters of Quebrada Bambú and Quebrada Fría represent a more dilute end member. Both long-term (over the 15 years) and short-term or seasonal decreases in concentration and steady or decreasing temperature are noted in NW springs. Springs located at the NE show increasing temperatures and ion concentrations, except for bicarbonate that has decreased in concentration for all the springs. This behavior is likely associated with a shallow source for the solutes and heat for this aquifer. To the NW the early lavas and pyroclastic flows have been cooling down, decreasing the contribution of leaching products to the infiltrating waters. To the NE, pyroclastic flows to the N during the last decade are contributing increasing concentrations of solutes and heat throughout water infiltration and circulation within the faults and the surficial drainage that has a NE regional trend. For the short-term or seasonal variations, concentrations of chemical constituents decrease when precipitation increases. However, correlations between concentrations and the number of seismic events per month and the modeled vertical tidal acceleration are also observed. The intrinsic periodic behavior of all these variables influenced by the Earth's rotation can complicate the interpretation of the chemical changes at hot springs. For example, variations in atmospheric pressure can affect the degassing and seismic tremor of a volcano as well as the rate of precipitation. Frequent monitoring and understanding of these variations is essential at every volcano if we want to use the variations in chemical composition of hot springs in volcanic monitoring.

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1. Introduction

Changes in the water chemistry of hot springs associated with volcanic activity have been observed throughout the world. At Popocatepetl Volcano, Mexico, changes in spring chemistry have been reported prior to main eruptions (Martin-Del Pozo et al., 2002). Species such as SO_4^{2-} , Cl^- , F^- , HCO_3^- , B and $\text{SO}_4^{2-}/\text{Cl}^-$ show changes in concentration. These species are related to the input of magmatic gases within hydrothermal systems surrounding the magmatic chamber. In volcanic gases, increases in the $\text{SO}_4^{2-}/\text{Cl}^-$ have been reported in several volcanoes such as Nevado del Ruiz (Williams et al., 1986), and Ruapehu (Giggenbach and Glover, 1975). In the hydrothermal system of Vesuvius volcano, Italy, changes in water chemistry were observed prior to a seismic event in October 1999 (Cinzia et al., 2004), particularly at Olivella spring at the northern flank of the volcano. A decrease in pH with corresponding increase in dissolved CO_2 as well as $^3\text{He}/^4\text{He}$ ratio was observed. In the springs of Popocatepetl (Martin-Del Pozo et al., 2002) as well as in Vesuvius (Cinzia et al., 2004), periodic variations in water chemistry are observed. For Vesuvius, deeper waters show more stable water compositions and shallower waters a higher amplitude of the variations suggesting stronger seasonal effects in the compositions.

Changes in water chemistry can be complicated by the influence of background processes in volcanic degassing. For example, the crater degassing and seismic tremor at Arenal Volcano has been correlated to Earth tides (Leandro and Alvarado, 1988; Williams-Jones et al., 2001). Correlation between volcanic seismicity and solid Earth tides have been found for other volcanoes of the world (e.g. Povlof Volcano, McNutt and Beavan, 1981; Kilauea Volcano, Rydelek et al., 1988). At Arenal, increasing tremor is observed when the rate of change of Earth tides is greater. A degassing mechanism very sensitive to small changes in the confining pressure has been suggested for this volcano. However, data relating seismic activity and Earth's tidal stress field have been examined for several volcanoes (Neuberg, 2000) including Stromboli in Italy, Mt. Ruapehu in New Zealand, and Merapi and Batur in Indonesia. Neuberg (2000) concluded that a comparison with theoretical body tides in the spectral domain show lack of evidence for the possible relationship between Earth tides and volcanic seismic activity. Neuberg states that other semi-diurnal parameters such as barometric pressure and temperature are probably better candidates to generate this phenomenon. For Stromboli, Neuberg

found a good spectral correlation between volcanic activity and barometric pressure. Barometric pressure and temperature also change seasonally, especially in Northern Costa Rica where two-season area is observed: rainy between April and December, and dry between January and March. The purpose of this paper is to explain the occurrence of the thermal springs within the volcanic structure of Arenal Volcano and to analyze their transient behavior in temperature and hydrochemical composition. Correlation with processes that could affect the water compositions and temperatures, such as precipitation, volcano seismicity, and Earth tides is investigated.

2. Geologic and hydrologic background

Arenal Volcano in Costa Rica (Fig. 1) has been the most active volcano of the Central American arc during the last 40 years with continuous activity since its explosive eruption in July 1968. It is considered one of the four most active volcanoes of the American continent (Simkin and Sieber, 2000) and one of the one hundred most active volcanoes of the world (Tazieff and Sabroux, 1983; Yokoyama et al., 1984). However, only a few hot springs of relatively low temperature are found in the area surrounding this volcano. Four hot springs with temperatures ranging from 38 to 61 °C (Fuente Doris (FD), Río Tabacón (RT), Quebrada Lava (QL), and Quebrada Guillermina (QG)) and two low temperature springs (23–29 °C, Quebrada Bambú (QB) and Quebrada Fría (QF)) have been sampled periodically during the last 15 years (Fig. 1). During the last decade, the Arenal Volcano region has been searched intensively looking for thermal springs for tourism development. As a consequence, three additional hot springs were identified (Laguna Verde (LV) and two at Río Arenal, which are called Montañas de Fuego (MF)) in Fig. 1. Two boreholes at Baldi Termale (B-1 and B-2), a tourist spa, have been sampled over the last 5 years.

Arenal Volcano is located between the Guanacaste and Central Mountain Ranges in NW Costa Rica (Fig. 1). It is a small stratovolcano with a 1660 m.a.s.l. and 1100 m elevation with respect to its base, and a 15 km³ volume (Alvarado and Soto, 2002). Its activity started around 7000 years ago. The present cycle of activity started in 1968 and has generated 0.7 km³ of lavas and pyroclastics (Alvarado et al., 2006-this volume). The products of this volcano are calc-alkaline basalt andesites low in potassium and rich in aluminum (Borgia et al., 1988). Arenal is located at the intersection of several Holocene active faults striking predominantly N–S, NW–SE, and NNW–ESE (Fig. 1). The location of the

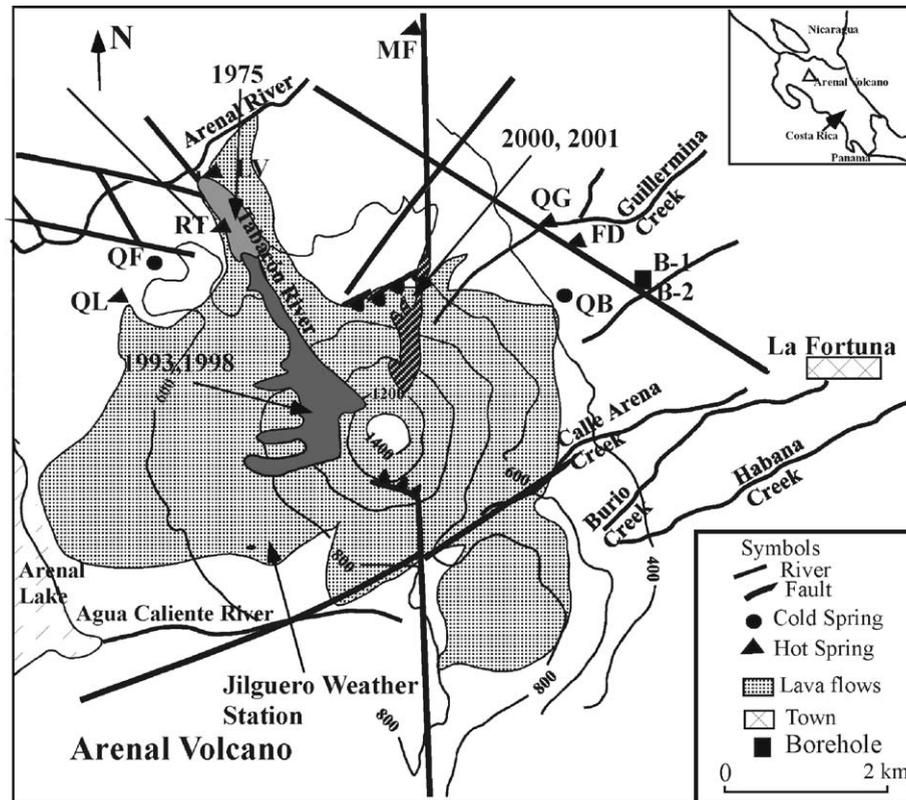


Fig. 1. Arenal Volcano in NW Costa Rica showing recent undifferentiated pyroclastic deposits (1995, 1998 and 2000, 2001), faults, streams draining the region, and the location of springs and boreholes. Faults with known sense of displacement are indicated with “-” and “+” sign (Soto and Sjöbohm, 2005). For the other faults, the sense of displacement is unknown.

known springs is clustered in the NW and NE quadrants, either related to contacts between the recent lava flows and older pyroclastic deposits (QL, LV, RT, QB) or to the faults in the area (RT, MF, QG, FD). At least one of these springs changed temperature before the present cycle of activity started. Río Tabacón hot spring showed an increase in temperature several months before the eruption of July 29, 1968 (Alvarado and Barquero, 1987). Few studies have been done in these springs and their relationship to the volcanic activity (e.g. Bigot and Barquero, 1986; Marini et al., 1990; Soto et al., 1995, 1999). Previous studies identified the water types and location of the springs within the volcanic setting. However, a detailed study of their transient behavior and chemical evolution has not been done.

Arenal Volcano is located in a tropical rain forest region with annual precipitation often higher than 5 m per year. The volcano is formed by a sequence of lava flows and pyroclastic deposits of relatively low primary permeability. Lava flows predominate to the east, and pyroclastic deposits interlayered with epiclastics and lava flows predominate to the west. The space between the blocky lava flows and fractures of the other deposits

provide high permeability flow paths for infiltration of rainwater and groundwater flow, making the edifice highly permeable. The contacts between the recent and older deposits seem to provide high permeability horizontal paths and the faults provide high vertical permeability, as can be inferred from the location of the springs in Fig. 1.

3. Methodology and database

Since late 1989, the four hot springs (RT, FD, QG, and QL) and two cold springs (QF and QB) have been sampled at approximately 3 month intervals, and at Baldi Termale (B-1 and B-2) from 2001 to 2005. The two boreholes have depth of, and are receiving water at 30 and 50 m, respectively. In this paper, we use data collected by Instituto Costarricense de Electricidad (ICE) from January 1990 to April 2005. Samples are collected in two 500 ml bottles for analysis of cations and anions. The cation samples are preserved with 5 N HNO₃ (1 ml per 100 ml). Field pH, conductivity, and temperature are measured in the field at the time of sampling. The water samples are analyzed for cations by

Atomic Absorption Spectrophotometer and for anions by titration methods (bicarbonate), spectrophotometric methods and ion chromatography. Errors in the determination of the different ions are lower than 10% in all cases. As the data collected during all these years is very extensive, only examples of early and late data are shown in Table 1 for November 1990 and November 2004.

The chemical compositions of Arenal springs were compared with the waters generated by interaction with average crustal rock (AC), average basalt (AB), average granite (AG), and a typical basalt andesite from Arenal (ABA). Composition of major cations for these rocks are presented in Table 2 as reported by Taylor (1964) for AC, AB, and AG, using data from around the world. For ABA data reported by Chiesa (1991) was used. 1 g of AC, AB, AG, and ABA was theoretically dissolved in 1 kg of water and the relative ionic composition of the generated waters was compared with Arenal springs. In June 1992, ICE collected samples for isotopic analysis of $\delta^{18}\text{O}$ and δD . These samples were analyzed at the Laboratory of Isotopic Hydrology of the OIEA in Vienna, Austria. Table 3 shows results for isotopes collected at that time. Reference values are the Standard Mean Ocean Water (SMOW), errors for $\delta^{18}\text{O}$ and δD determinations are $\pm 0.08\text{‰}$ and $\pm 1\text{‰}$, respectively.

Monthly precipitation at the closest weather station to Arenal Volcano (Station Jilguero, Fig. 1) for the time period 1990–2004 was provided by the Center for Basic Engineering Studies of ICE. Note that atmospheric pressure is not measured in this station. Data for the

Table 2

Concentration (mg/kg of rock) for average crustal rock, average basalt, and average granite (Taylor, 1964) and average Arenal basaltic andesite (Chiesa, 1991)

	Initials	Na	K	Ca	Mg
Average crustal rock	AC	23,600	20,900	41,500	23,300
Average basalt	AB	19,400	8300	67,200	45,000
Average granite	AG	15,800	1600	27,700	33,400
Arenal average basaltic andesite	ABA	22,400	4700	66,200	23,900

seismicity of Arenal Volcano was provided by Observatorio Sismológico y Vulcanológico del Arenal y Miravalles (OSIVAM, ICE) as they recorded it in two analog stations located at Arenal Volcano: Chiripa (AR6) and Fortuna (FOR), during the period 1990–2002. The number of explosive seismic events per month was used in this investigation.

We used the program Etagtab v. 3.0 (Wenzel, 1996) to calculate the vertical component of the tidal acceleration of gravity at Arenal and to investigate the effect of Earth tides on seismicity and precipitation during the study period (1990–2004).

4. Results and discussion

4.1. Chemical composition

According to Giggenbach (1984, 1988), the waters discharging from a volcanic hydrothermal system can

Table 1

Analytical results for pH, temperature ($^{\circ}\text{C}$), Na^+ , K^+ , Ca^{2+} , Mg^{2+} , SiO_2 , Mn^{2+} , B, Cl^- , NO_3^- , SO_4^{2-} , F^- , and HCO_3^- (mg/l) for each spring and borehole

Site	Initials	pH	T ($^{\circ}\text{C}$)	Na	K	Ca	Mg	SiO_2	Mn	B	Cl^-	NO_3^-	SO_4	F	HCO_3^-
<i>November 1990</i>															
Fuente Doris	FD	6.05	35.9	95.1	15.6	54.4	50.3	98.9		0.95	81.1	3.26	147.0	0.21	301
Quebrada Fria	QF	6.05	26.3	67.7	9.8	103.0	76.4	86.2		0.67	217.0	1.23	35.4	0.17	449
Quebrada Bambú	QB	6.05	22.4	9.6	2.5	17.6	4.7	75.1	0.03	0.05	4.2	1.17	1.7	0.08	102
Quebrada Lava	QL	7.25	49.7	259.0	37.9	111.0	34.5	110.0	3.17	2.22	570.0	4.10	29.6	1.20	203
Quebrada Guillermina	QG	8.10		16.0	3.7	25.2	15.6	73.6		0.13	7.2	0.68	24.8	0.27	160
Rio Tabacon	RT	6.10	46.3	303.0	28.5	140.0	107.0	106.0	3.55	2.16	609.0	12.30	303.0	0.41	395
<i>November 2004</i>															
Fuente Doris	FD	6.41	39.4	138.0	23.4	72.8	55.8	92.0	1.36	1.53	231.0	9.10	230.0	0.23	207
Quebrada Fria	QF	6.62	30.5	48.3	7.0	53.0	43.9	85.9	0.03	0.38	78.0	2.82	56.0	0.21	333
Quebrada Bambú	QB	6.70	24.7	10.0	2.2	12.9	3.2	69.9	0.03	0.09	6.3	0.66	2.9	0.17	81
Quebrada Lava	QL	7.08	32.6	35.2	4.5	10.4	4.8	69.9	0.06	0.23	25.7	0.32	29.2	0.91	84
Quebrada Guillermina	QG	6.35	42.6	178.0	27.8	87.7	64.5	106.0	0.10	2.08	324.0	14.30	369.0	1.20	162
Rio Tabacon	RT	6.58	49.5	213.0	27.9	82.5	54.6	150.0	0.06	1.49	337.0	4.28	296.0	0.18	229
Laguna Verde	LV	6.87	48.5	113.0	19.7	34.7	11.7	98.6	7.73		130.0	2.64	51.7	0.47	216
Montana de fuego	MF	8.44	34.5	206.0	31.3	77.7	69.9	140.0	0.08	2.65	378.0	0.54	1.7	0.07	840
Borehole B-1	B-1	7.93	38.0	102.0	13.3	146.0	56.5	83.9	0.03	1.13	225.0	1.67	259.0	0.07	289
Borehole B-2	B-2	7.47	49.5	244.0	39.1	106.0	91.1	114.0	3.31	2.76	429.0	3.52	214.0	0.31	263

Table 3
Isotopic composition of Arenal Volcano springs

Spring	Initials	δD	$\delta^{18}O$
Quebrada Bambú	QB	-27.1	-4.92
Fuente Doris	FD	-27.1	-4.70
Río Tabacón	RT	-20.0	-2.62
Quebrada Fría	QF	-27.3	-4.51
Quebrada Lava	QL	-22.4	-3.72

be classified according to their anionic composition (Cl^- , SO_4^{2-} , HCO_3^-). Fig. 2 shows the triangular diagrams for the anionic composition of the six springs of Arenal and the boreholes B-1 and B-2 for the many sampling events during the study period. Quebrada Lava (QL), Río Tabacón (RT), and Quebrada Fría (QF) were chloride waters at the beginning of the study period. RT did not evolve considerably during the time of this study (Fig. 2B). However, the relative HCO_3^- concentrations at QL and QF increased over time. Quebrada Bambú (QB) remained a bicarbonate water over the study period. The relative concentrations of Cl^- and SO_4^{2-} increased over time at Quebrada Guillermina (QG) and Fuente Doris (FD), respectively. For B-1 and B-2, the position in the diagram fluctuated around the small region shown in Fig. 2F and G. As was noted by Soto et al. (1995, 1999), the springs with higher relative Cl concentrations (Table 1) were those located to the NW of the volcano (RT, QL, QF), and those located to the NE had higher relative bicarbonate concentrations (FD, QG, QB).

Scatter plots of Cl versus B and K versus Na for November 1990 and November 2004 show linear trends for each date (Fig. 3) suggesting a single aquifer or waters generated by a similar chemical processes (e.g. the dissolution or alteration of Arenal volcanic rocks). Note that for Cl and B, the linear trend changed from 1990 and 2004 towards increasing B content, reflecting an increased contribution of this element to the aquifer. Based on the linear trends observed in Fig. 3A and B, the waters can be interpreted to reflect the mixing of two end-member waters, one more saline with the composition of Río Tabacón (RT) to the NW, and another diluted water with the composition of QB in 1990. In 2004, the end member for the saline water seems to have changed to the composition of borehole B-2 to the NE.

For the cations, the linear trend changed from 1990 to 2004 towards increased potassium concentration (Fig. 3B). The waters of Arenal Volcano are very rich in Mg compared to Na and K. They fall in the extreme Mg corner when a triangular diagram of Na/1000, K/100,

and $Mg^{1/2}$ (Giggenbach, 1988) is plotted, which is interpreted as waters with minimal water–rock interaction. This condition is also reflected in the diagram for the equilibrium of the mineral assemblage albite, K-feldspar, muscovite, clinocllore, calcite, and quartz (Giggenbach, 1988) (Fig. 4). In this diagram, the compositions of the Arenal waters, and the waters generated by the dissolution of average crustal rock (AC), average basalt (AB), average granite (AG), and a typical Arenal basaltic andesite (ABA) are plotted. The waters fall close to the ABA dissolution zone far from the equilibrium line indicating an early stage of rock–water interaction.

The isotopic compositions of the Arenal waters in Fig. 5 (Soto et al., 1995) show that they originate as the mixing of andesitic magmatic water (Giggenbach, 1992) and meteoric water as described by Craig (1961). Considering the mixing of these two waters, a fraction of 20%, 10%, and 2% of andesitic water is found for RT, QL, and FD.

4.2. Transient variations in composition

Chemical data collected over 15 years for springs of Arenal Volcano offer an opportunity to study the transient variations in water composition in the edifice of an active volcano. Plots of concentrations of chemical species versus time show clear increasing or decreasing long-term trends (over 15 years) as well as short-period (or seasonal) variations (Figs. 6–9). The water temperature of all springs increased by a few degrees over the study period, except in Quebrada Lava where the temperature dropped from 61 to 32 °C, and at Río Tabacón (RT) and the two boreholes B-1 and B-2, where the temperature is variable but does not show a significant long-term average change. Average pH increased slightly (tenths of unit) at all springs, except at Fuente Doris (FD), B-1, and B-2, where it remained more stable. In general, springs located on different flanks of the volcano have distinct decreasing or increasing long-term concentration trends. For the ions Na, K, Ca, Mg, sulfate, and chloride (and B with high noise), springs located to the NW have decreasing concentration trends (RT and QL are shown in Figs. 6 and 7 as examples) and springs located to the NE have increased concentrations with time (FD is shown in Fig. 8 as example). The ratios SO_4^{2-}/Cl^- have an opposite behavior with a decreasing trend to the NE and an increasing trend to the NW. The decreasing concentration long-term trends for QL are more intense than for RT and QF. Na concentration has dropped from more than 200 to less than 50 mg/l.

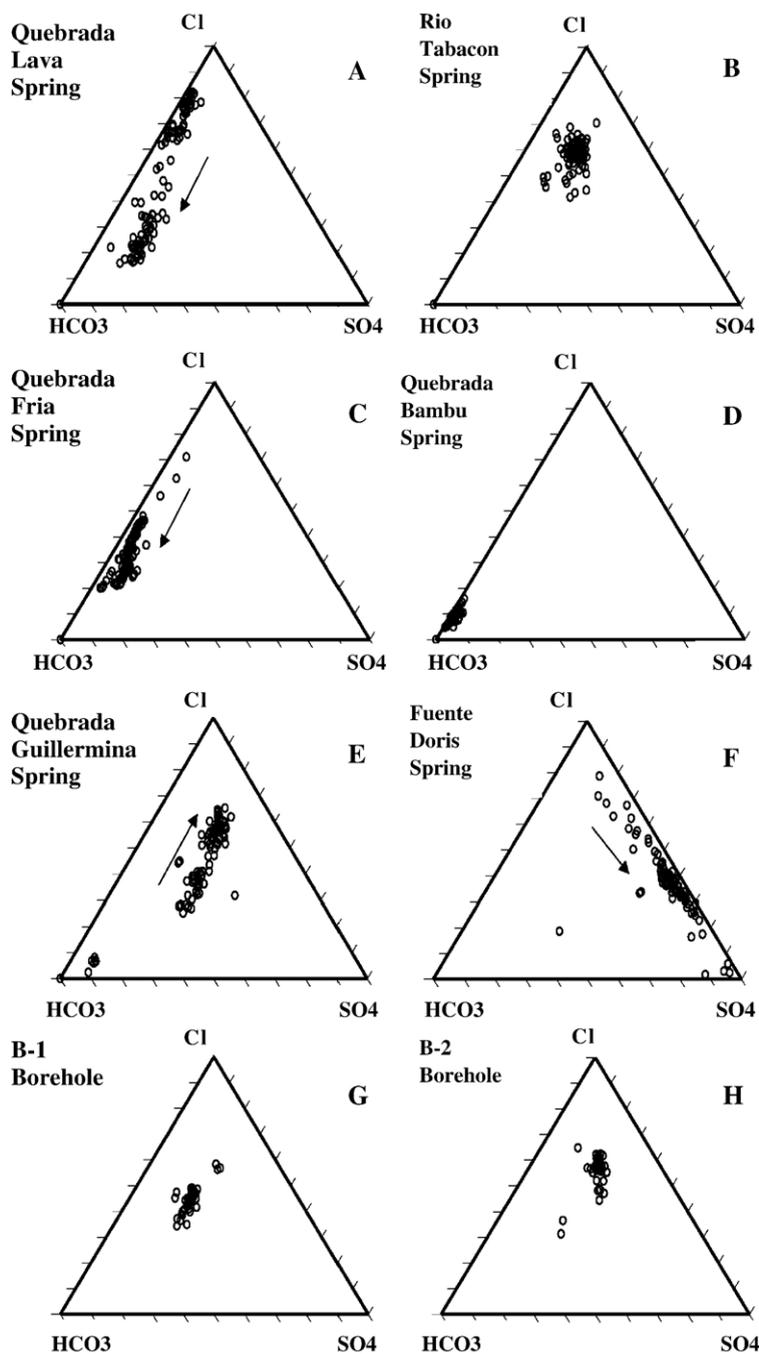


Fig. 2. Triangular diagram (HCO_3^- , Cl^- , SO_4^{2-}) for hot springs of Arenal Volcano that were sampled frequently from 1990 to 2005. Also shown are B-1 and B-2 which were sampled from 2001 to 2005. Arrows indicate evolution of the water composition from 1990 to recent.

K has decreased from more than 45 to around 10 mg/l, Ca from more than 120 to less than 20 mg/l, Mg from around 50 to less than 10 mg/l, sulfate has stayed stable, but Cl has decreased from around 600 to a few mg/l. The increases in concentration in the springs of the NE are equally important. Na and K in FD have

almost doubled (Fig. 8B) as well as Cl (Fig. 8D), Mg has increased from around 45 to around 65 mg/l and Ca from around 110 to around 150 mg/l. Again, only slight changes in long-term sulfate concentrations are observed. As expected the ratio $\text{SO}_4^{2-}/\text{Cl}^-$ has increased.

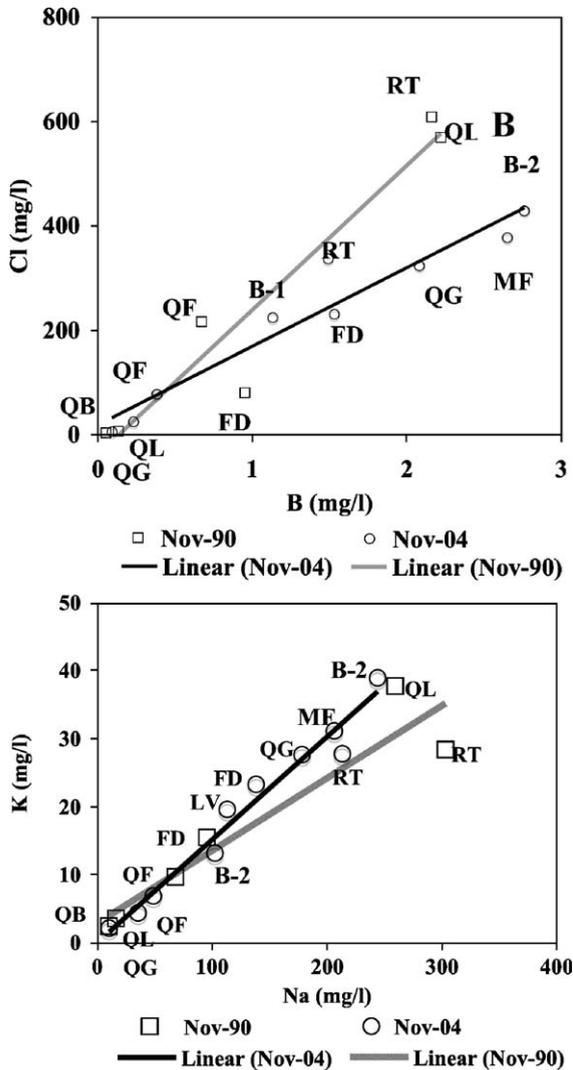


Fig. 3. Plots of Cl versus B, and K versus Na for the thermal waters of Arenal Volcano. Squares represent 1990 data and circles 2004 data.

In our previous work, López et al. (1994) and Soto et al. (1995, 1999) suggested that the hot springs of Arenal have two possible sources of magmatic chemical species. One is the internal magmatic body that can contribute chemical species to the hydrothermal system surrounding the magmatic environment. Faults and fractures within the volcanic edifice can provide fast permeable paths for magmatic gases to migrate to the shallower aquifers. The other source is the hot lava flows recently erupted by the volcano as well as the deposition of erupted products from the atmosphere. Infiltration of water into the hot materials can provide the right environment for alteration reactions and transport of chemical species to the discharging springs. Quebrada Lava (QL) is located at the contact between

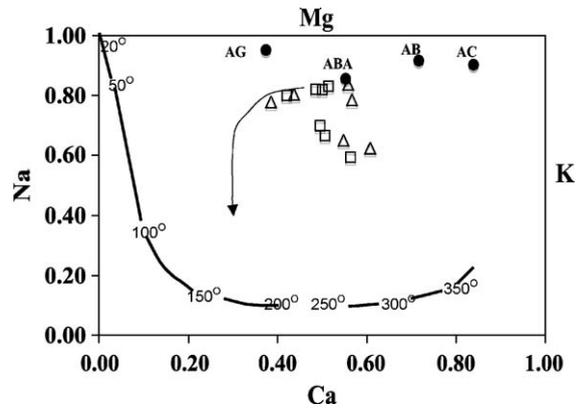


Fig. 4. Waters of Arenal Hot Springs plotted in a diagram of $(10Mg/(10Mg+Ca))$ versus $(10K/(10K+Na))$ showing that the waters of Arenal fall close to the region of dissolution of Arenal basaltic andesites (ABA). The triangles represent waters from November 1990 and the squares, waters formed by the dissolution of 1 g of average crustal rock (AC), average basalt (AB), average granite (AG), and a basaltic andesite from Arenal Volcano (ABA) in 1 l of water. The arrow indicates the path that the water could follow to reach mineral equilibrium as described in Giggenschbach (1988).

the recent lava flows and the older volcanic rocks. Río Tabacón (RT) is located at one of the faults that intersect the volcano, close to the 1975 pyroclastic flow (Fig. 1). QL is probably receiving more water that circulates throughout the recent lava flows than RT. With time, the lava flows have been cooling down providing less heat to the circulating water (as indicated by the decrease in temperature at QL) and less chemical species because of lower reaction rates associated with decreasing temperature. RT shows a similar effect. However, as the

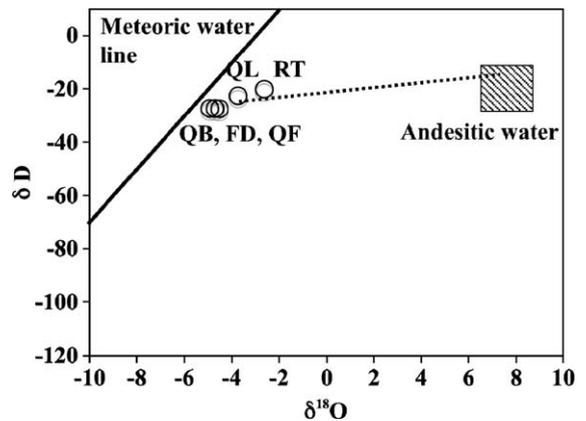


Fig. 5. Isotopic composition of Arenal Volcano hot springs. The more diluted waters of Quebrada Bambú (QB), Fuente Doris (FD), and Quebrada Fria (QF) fall close to the meteoric water line. Río Tabacón (RT), and Quebrada Lava fall close to the water mixing line between the meteoric water line and the andesitic water as defined by Giggenschbach (1992).

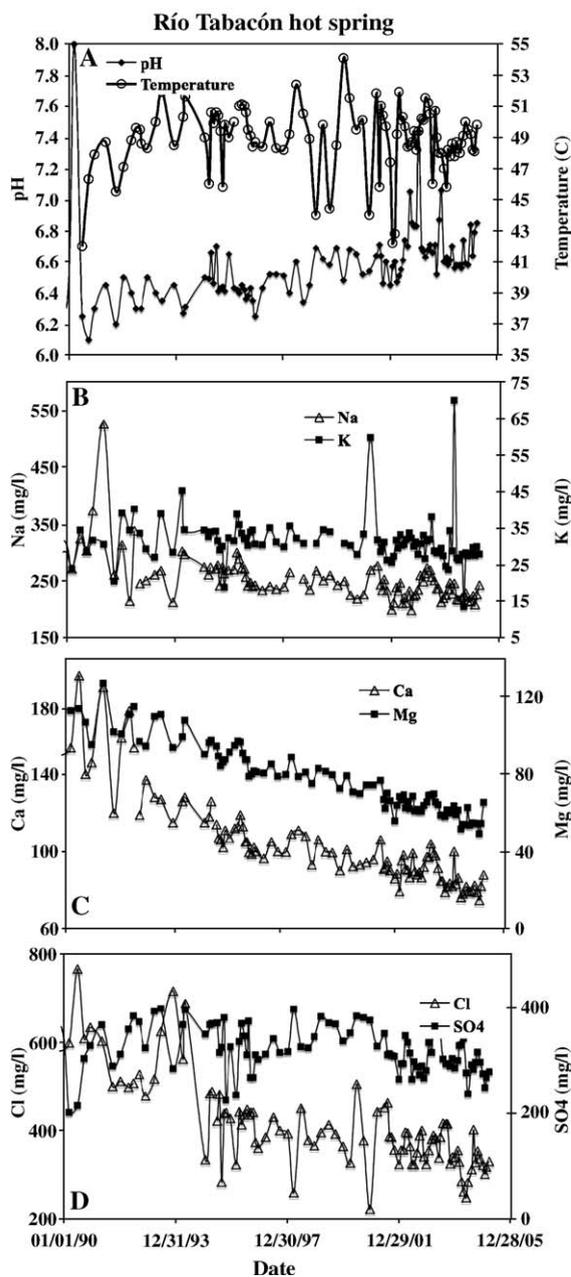


Fig. 6. Variations of chemical composition from 1990 to 2004, Río Tabacón hot Spring, Arenal Volcano. Trends of increasing pH and decreasing concentrations with time are observed.

proportion of meteoric water in this spring is lower, it receives more water from the deeper circulation system. A higher fraction of deep water produces a milder decrease in concentration. Temperature is more stable at RT, likely reflecting a more direct connection to the deeper system. QF is also located along a fault that intersects the volcano. The composition of this spring is highly diluted by meteoric water, but the spring is not

located at the contact with the recent lava flows. For that reason, its behavior is more similar to RT than to QL.

The two boreholes B-1 and B-2 also have short-term variations in pH, temperature and concentrations (e.g. Fig. 9 for B-1). However, pH and temperature do not show a significant average increase over time (e.g. Fig. 9A). For the anions and cations, B-1 presents a trend similar to the NE springs but with a milder increasing slope (except for HCO_3^- which decreases). In comparison, concentrations of the different chemical species for B-2 have fluctuations but a definite overall increasing or decreasing trend is not observed. This behavior in water chemistry at the two boreholes is consistent with the

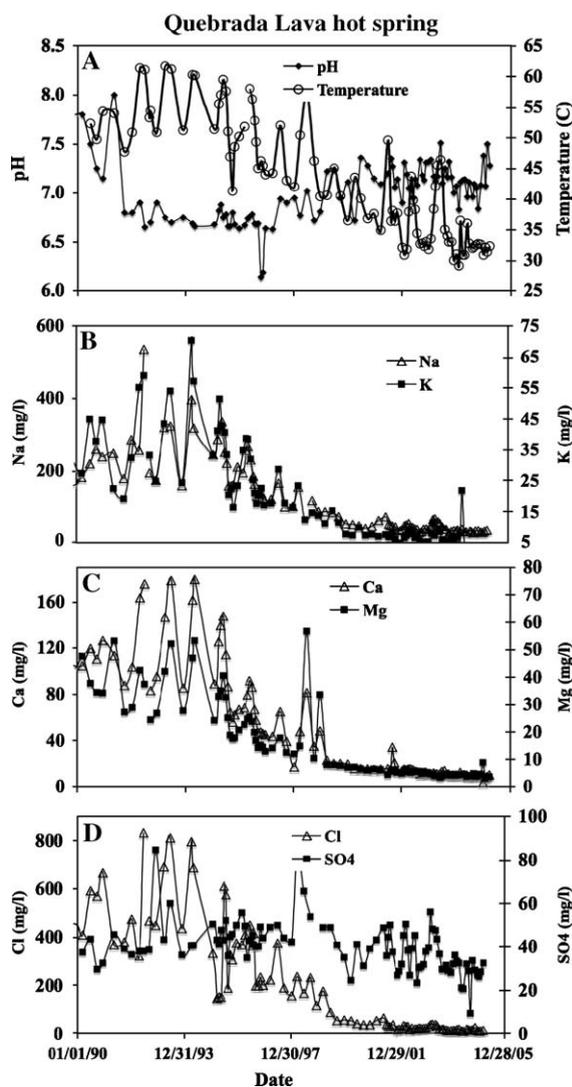


Fig. 7. Variations of chemical composition of Quebrada Lava hot spring from 1990 to 2004. Trends of decreasing temperature and concentrations with time are observed.

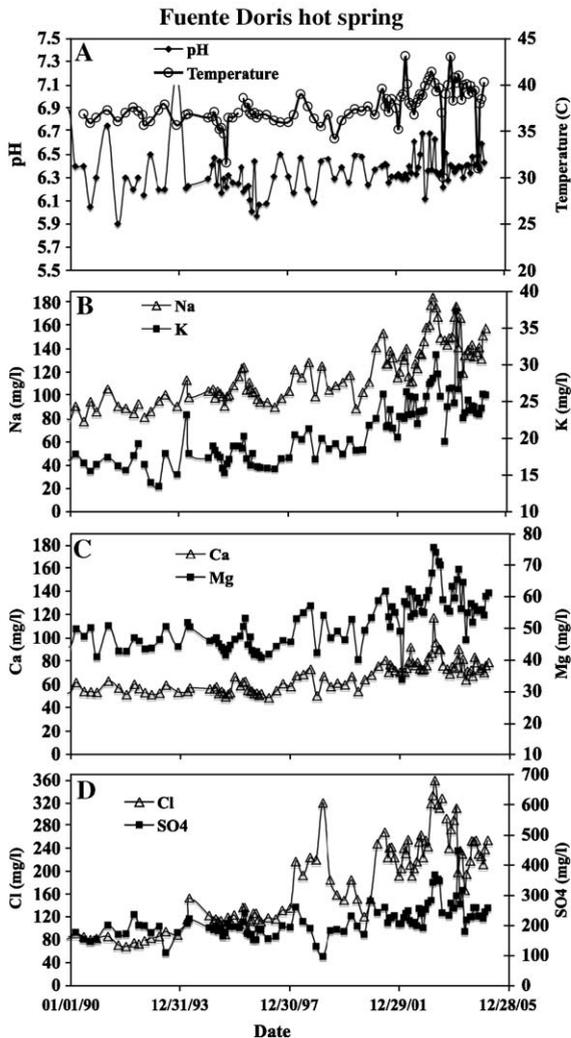


Fig. 8. Variations of chemical composition of Fuente Doris hot spring from 1990 to 2004. Trends of increasing temperature and concentrations with time are observed.

lower temperature observed at B-1 (around 40 °C) compared with the temperature at B-2 (around 50 °C). These values of temperature suggest a higher mixing ratio of cold meteoric water for B-1 than for B-2, as it should be expected because B-2 is slightly deeper. As in Vesuvius Volcano (Cinzia et al., 2004), transient changes in water chemistry are probably related to the circulation depth of the water reaching the wells.

The long-term increase in concentrations in the springs located to the NE probably reflects a different process. QG, B-1, B-2 and FD are both located along the same fault. QB is close to a contact but it is the most diluted water reflecting low water–rock interaction. The increase in concentration of Cl and sulfate in the NE springs as well as the concentration of cations, in

addition to increases in temperature, suggest that they could be receiving higher heat fluxes in 2004 compared with 1990 and probably more magmatic components. However, when the concentration of bicarbonate versus time is plotted for all different springs including the NW springs, a strong decreasing long-term trend can be observed (Fig. 9D for B-1, Fig. 9 for RT, QL, FD, and QG as examples; Fig. 10). The increasing trend in pH for all these springs and decreasing trend for bicarbonate are contradictory if a constant partial pressure of CO₂ in

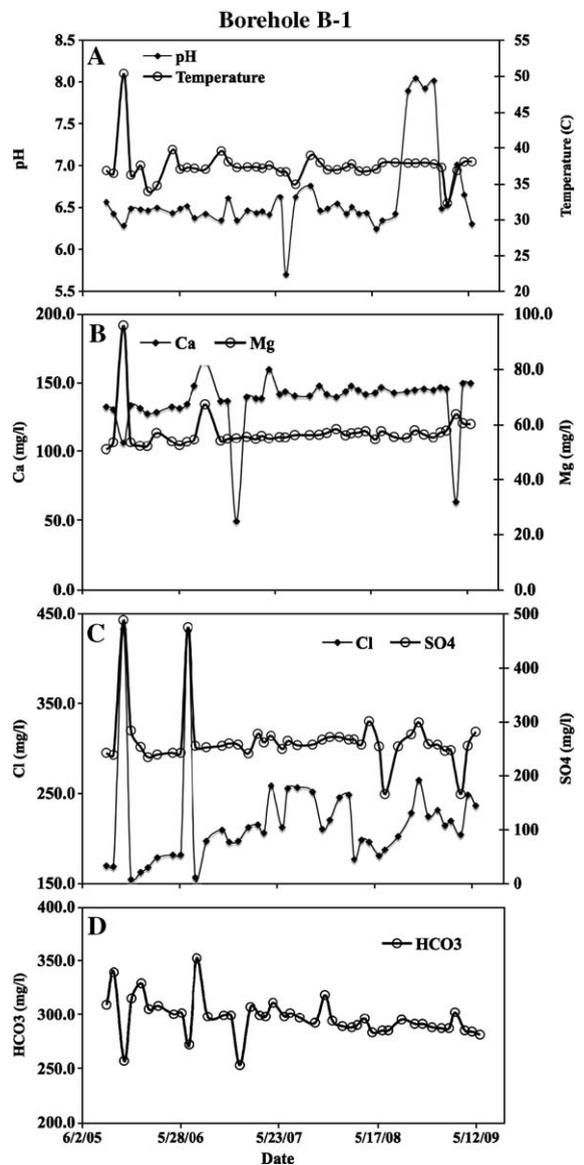


Fig. 9. Variations of chemical composition of borehole B-1 from 2001 to 2005. pH and temperature oscillate but the average trend is stable. A mild increase in concentrations is observed except for bicarbonate that decreases.

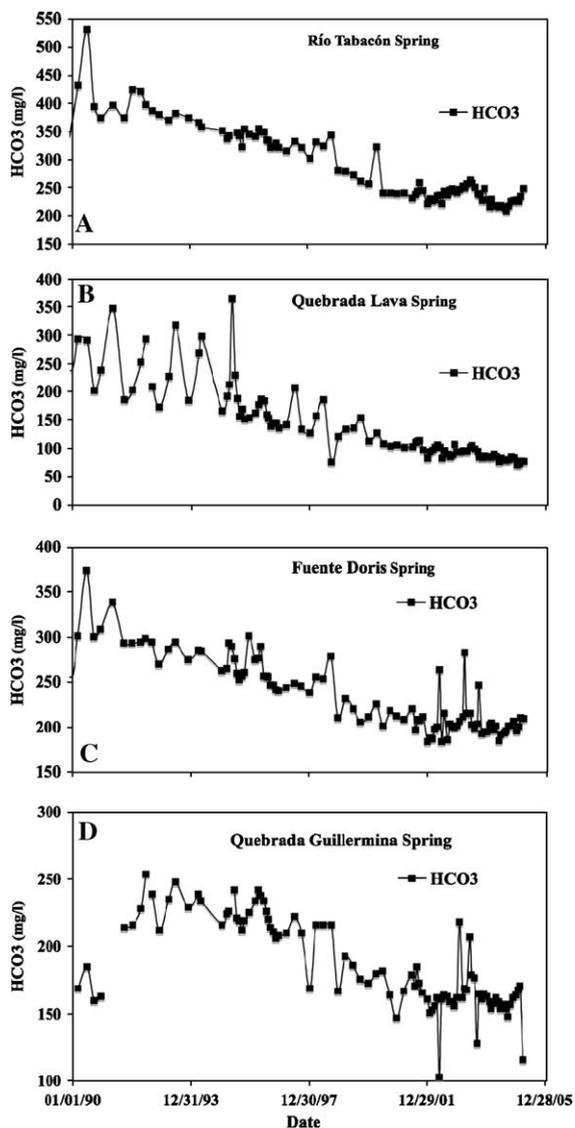


Fig. 10. Concentrations of bicarbonate versus time for the springs of Arenal Volcano. A decrease trend in HCO_3^- concentration with time is observed in all the springs.

the aquifer is considered. The concentration of bicarbonate depends on both the partial pressure of CO_2 (P_{CO_2}) and the concentration of hydrogen ions (a_{H^+}):

$$m_{\text{HCO}_3^-} = K_1 K_{\text{CO}_2} P_{\text{CO}_2} / a_{\text{H}^+}$$

(Drever, 1997) where K_1 and K_{CO_2} are the first dissociation constant for carbonic acid and the Henry's law constant for CO_2 , respectively. A decrease in the concentration of bicarbonate can be generated by a decrease in pH or a decrease in the P_{CO_2} . A decrease in pH has not occurred. On the contrary, pH has increased

slightly in almost all the springs. As a consequence, the P_{CO_2} in the aquifer should have decreased, which means that magmatic inputs of gases to the hydrothermal system have probably decreased over time. This is indirectly shown in the volcanic activity, where lava flows have decreased their length and extrusion rate since 1998, suggesting a lower rate of magma feeding.

If the source of the increase in the concentration of chemical species, especially Cl and sulfate, at the NE springs of Arenal is not the internal source of magmatic gases, we are left only with the possibility of the external or shallower source. It is possible that the more recent deposits are providing Cl, sulfate and cations as leaching of the hot materials occurs in a similar way as it happened at Quebrada Lava at the beginning of the study period. In fact, since 1998, the presence of spatter cones and lava outflow moved to the northern part of the active crater. Also, lava front rockslides and pyroclastic flows occurred more frequently on the northern slope, contributing juvenile material prone toward leaching, which likely provided an increase in dissolved chemical constituents to the aquifer. The number of pyroclastic flows increased significantly at Arenal since 1990 (particularly 1992–1993, 1996 and 1998), with the most notorious examples occurring in 1993 (flanks W and NW), 1998 (flank NW), 2000 and 2001 (flank N) (Alvarado and Arroyo, 2000; Alvarado and Soto, 2002) as shown in Fig. 1. Immediately after the deposition of the pyroclastic flow of October 23, 2000, ephemeral and small solfataras were formed. Sampling and identification of minerals deposited in these solfataras showed the presence of highly soluble salts, particularly salmiac (NH_4Cl) (Alvarado, personal communication). The presence of this salt can explain the increase in Cl observed in the NE springs. Note that the pyroclastic flows have been extruded to the NW and N flanks but the general surficial drainage of the area is towards the NE (Fig. 1). The shallow circulation of the ground water probably follows the same path. Also present are faults striking to the NE. Transport of leached chemical species and heat throughout faults and surface drainage could explain the increase in temperature and chemical constituents of the NE springs.

4.3. Short-term variations in composition, seismicity and Earth tides

The short-term or seasonal variations in temperature and chemical composition observed in all the waters and for all the ions at the springs of Arenal can be correlated with the precipitation or with inputs of magmatic gases to the hydrothermal system. To investigate the possible

correlation of precipitation with the variations of chemical composition, the concentrations and monthly precipitation versus time were plotted. Mg is used as an example in Fig. 11 for QG, QL, QF, and FD. As the chemical data was collected in average every 3 months, correlation between rainfall and precipitation could be observed only at the frequency of seasonal fluctuations. In the Arenal area, two seasons (dry and rainy seasons lasting approximately 4 and 8 months) occur. For that reason a 6 month moving average trend was applied to the monthly precipitation to eliminate shorter frequency noise. As shown in Fig. 11, precipitation and concentration of chemical constituents in the different springs behave as periodic variables, with higher concentrations at the times that we have lower precipitation. For the relationship between two periodic variables, a cross-correlogram can be constructed instead of just finding a singular correlation coefficient. However, as the frequency of collection of the chemical data is low and the time intervals between successive sampling events are not uniform, it is not possible to identify a clear lag between the two variables, if it exists. A basic condition to construct a cross-correlogram is that the time intervals between sampling events must be uniform (Box et al., 1994). Only a qualitative study of the relationship between precipitation and concentrations of chemical species is possible at the present time.

One possible explanation of the relationship between precipitation and concentrations is that infiltration could be diluting the spring waters. Water infiltrating the young volcanic products interacts with the rock minerals and generates springs with high influence of the infiltrating water and with compositions consistent with the lack of chemical equilibrium observed in Fig. 4. For the groundwaters of Vesuvius (Cinzia et al., 2004) the amplitude of seasonal fluctuations is indicative of the depth of circulation of the discharging water. If the observations of seasonal fluctuations in springs and wells of Vesuvius apply to Arenal waters, the springs of Arenal are discharging waters with large proportion of fast moving shallow infiltration water.

Due to the low frequency of water sampling at Arenal springs, it is not possible to observe the direct effect of specific rainfall events on the water chemistry. Precipitation, at least for the first rainfall events of the raining season, could significantly increase the concentration of dissolved compounds in the aquifer. The reason is the dissolution of highly soluble compounds from the volcanic ash and the products of oxidation reactions stored during the dry season in the superficial

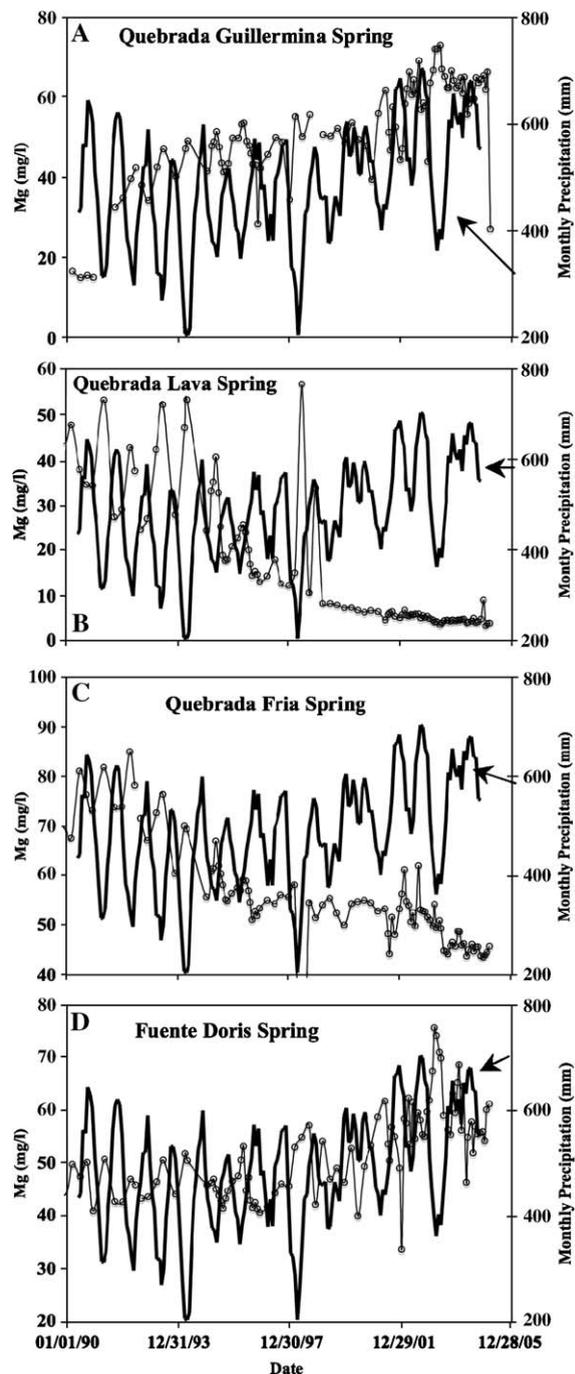


Fig. 11. Monthly precipitation and Mg concentrations in Arenal Volcano hot springs from 1990 to 2004. The 6 month moving average of precipitation is plotted. Heavy periods of rainfall produce dilution effects.

deposits. In other aquifers, such as the aquifers affected by acid mine drainage from coal exploitation in Southeastern Ohio, this effect is observed during the high flows of early spring (López and Stoertz, 2001).

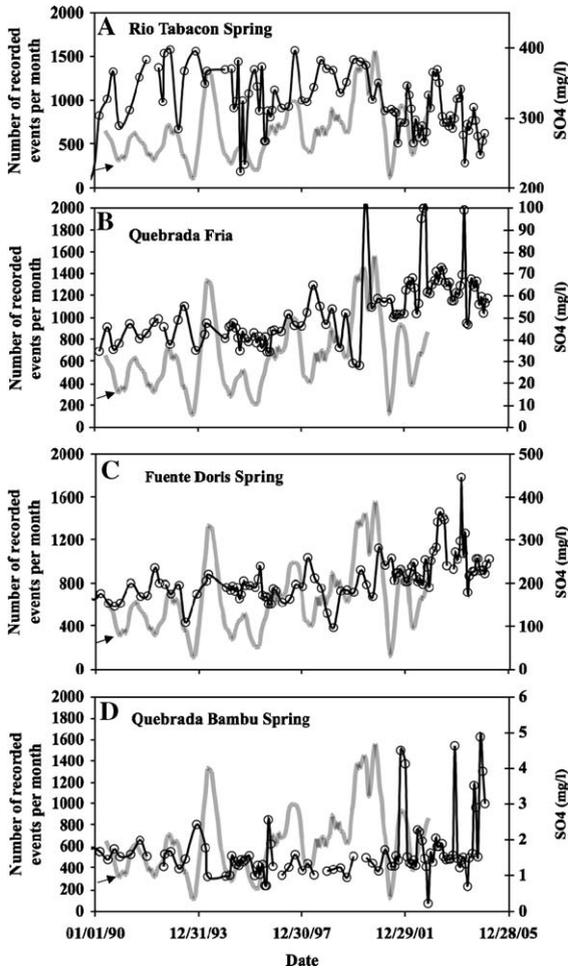


Fig. 12. Variations in sulfate concentration of hot springs and seismicity at Arenal Volcano. The 6 month moving average for the monthly number of earthquakes is shown.

As the rainfall season continues a net dilution effect is observed due to the high volume of water circulating throughout the reaction sites. This dilution effect is probably more important in Arenal due to the high annual precipitation.

A good indicator of contributions of magmatic gases to the aquifer could be a correlation with the explosive seismicity of the volcano. During seismic events inputs of gases to the hydrothermal system could occur. To investigate this possibility the number of seismic events per month and the concentration of sulfate (volcanic gases input sulfur gases to hydrothermal systems, Giggenbach, 1980) were plotted versus time (RT, QF, FD, and QB are shown as examples in Fig. 12). The 6 month moving average of the number of seismic events was plotted. Again, correlation between the fluctuations in the number of seismic events and concentration was observed. In this case, a small lag

between the two time series is observed. In general, the peaks in seismic activity occur close to the peaks in concentration suggesting possible connection between the two variables, as it has been reported in Popocatepetl Volcano (Martin-Del Pozo et al., 2002). However, this is contradicted by the lack of correlation between seismic activity and HCO_3^- concentration or temperature at Quebrada Lava (QL). As concentration seems to be correlated with precipitation and seismicity, precipitation and seismicity should also be correlated. As we have two different seasons (dry and wet seasons) in Costa Rica, lasting between 4 and 8 months, Fig. 13A shows the 6 month moving average of these two variables plotted against time. The peaks and valleys of these two functions seem inversely correlated at least from the 1990 to 1997 period, shifting to a positive correlation of peaks after that time. Fig. 13B and C explore the correlation between the variations in vertical tidal acceleration and the number of seismic events and monthly precipitation, respectively, for the studied period. In these two graphs the cyclic nature of the three variables is reflected. The vertical tides and precipitation have obvious annual

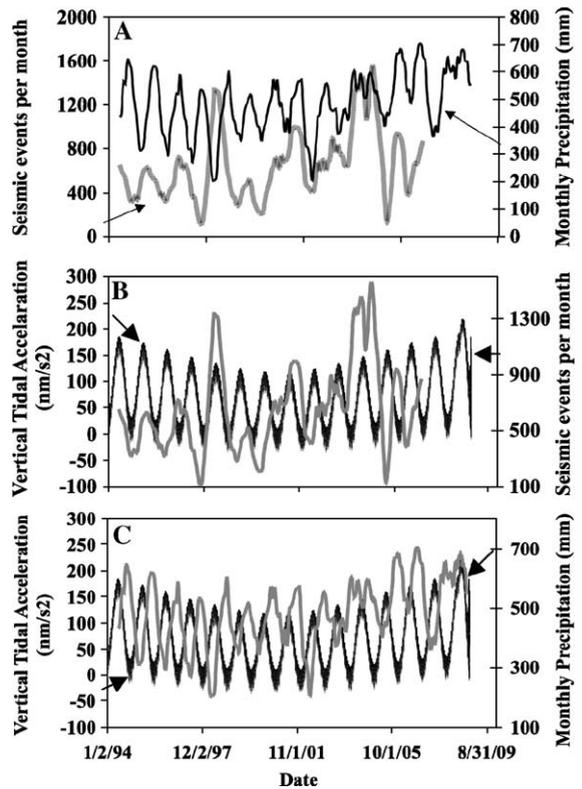


Fig. 13. Number of seismic events per month and monthly precipitation compared with Earth tides (vertical acceleration of gravity). The 6 month moving average of the three variables are plotted.

cycles that correspond to the movement of the Earth. Seismicity, on the other hand, could be the result of magma degassing affected by variations in the pressure field generated by the Earth tides, or as suggested by Neuberg (2000), the result of changes in atmospheric variables such as atmospheric pressure that promote degassing along the volcano conduit and consequent seismic tremor. If this is the case, as precipitation is also a function of atmospheric conditions, the relationships between all these variables could be the result of their intrinsic annual cycles.

4.4. Spectral analysis and cross-correlations of monthly precipitation, seismicity and Earth tides

A better understanding of the relationship between monthly precipitation, seismicity and vertical tidal acceleration can be obtained using autocorrelograms, cross-correlograms, and the spectral density functions of these variables. To construct an autocorrelogram, correlation coefficients are obtained comparing the variable with itself but shifting the values with respect to each other several lag times (Swan and Sandilands, 1995). The autocorrelogram is the graph of the correlation coefficients versus the lag time. A cross-correlogram is similar to an autocorrelogram, but in this case the lagged variable is a different variable. One condition for this kind of diagram is that the time intervals between sampling events have to be uniform. Monthly averages for the three variables were used in our analysis. The spectral density as a function of the frequency provide information about the dominant frequencies in the data set (Box et al., 1994). Fig. 14A show the autocorrelograms for the number of seismic event per month, the average vertical tidal acceleration, and monthly precipitation. The periodic nature of these variables is observed in the cyclic correlation coefficients. The vertical tidal acceleration presents the strongest periodicity (higher correlation coefficients), followed by precipitation, and seismicity with lower correlation coefficient values. Note the coincidence of the precipitation and vertical tidal acceleration peaks. The spectral density functions (Fig. 14B) show two well-defined peaks for vertical tidal acceleration and precipitation at 0.083 and 0.166 month^{-1} , which correspond to 12 and 6 month periods, as expected from the known annual variations of these variables. The monthly number of earthquakes or seismicity presents several peaks with two close to the 0.166 month^{-1} frequency peak and two close to the 0.083 month^{-1} peak. Finally, the cross-correlograms (Fig. 14C) between the pairs of variables (vertical tidal

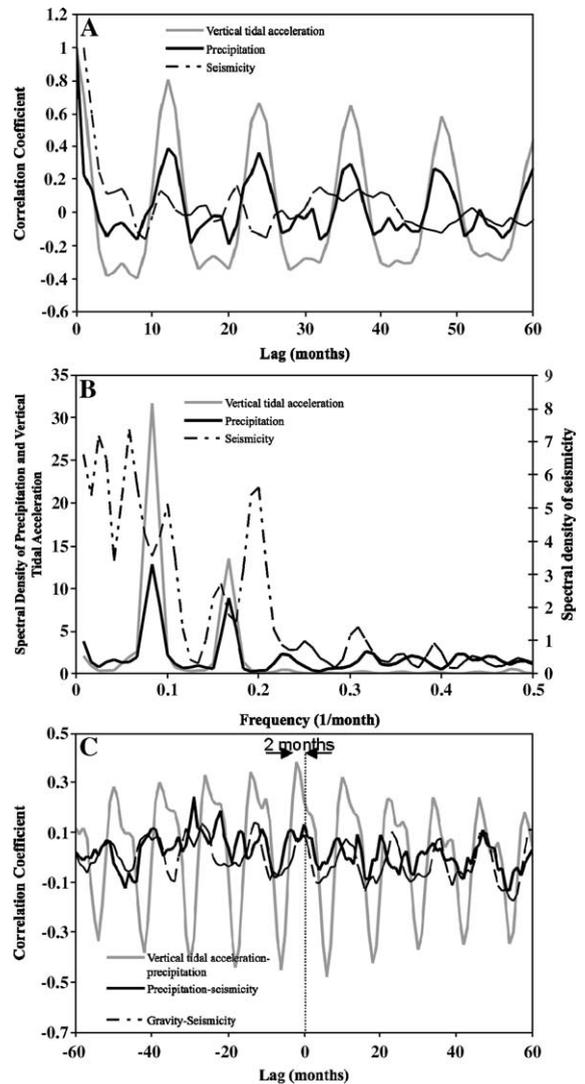


Fig. 14. Spectral and cross-correlation analysis of precipitation, seismicity, and vertical tidal acceleration at Arenal Volcano. (A) Autocorrelograms of the three variables showing their periodicity. (B) Spectral density functions showing peaks at frequencies of 0.083 and 0.166 month^{-1} for precipitation and vertical tidal acceleration and close peaks for seismicity. (C) Cross-correlograms for the different pairs of variables. A lag of 2 months for vertical tidal acceleration and precipitation is observed, and zero months for precipitation and seismicity, and vertical tidal acceleration and seismicity.

acceleration and precipitation, precipitation and seismicity, and vertical tidal acceleration and seismicity) clearly show how all these variables have periodic behavior and are correlated. Precipitation and seismicity, and vertical tidal acceleration and seismicity present maximum values at zero lag. Vertical tidal acceleration and precipitation present a lag of 2 months but the strongest cross-correlation. It is clear from these diagrams that the three variables present similar

periodicity and short lags, at least in the long period range considered here (months and higher).

5. Conclusions

The short-term variations in chemical composition in the waters of Arenal Volcano could be linked to the sources identified previously: circulation of infiltrating water within the hot volcanic products, or sudden inputs of volcanic gases to the aquifer due to changes in degassing conditions. These changes in degassing conditions can either be generated by variations in Earth tides or, more likely, variations in atmospheric conditions that are transferred throughout the volcanic conduit. Arenal has an open chimney that connects the atmosphere with the magma chamber, as evidenced by the frequent emission of volcanic products during more than 30 years. Changes in atmospheric pressure can be transferred to the magmatic chamber affecting the degassing process. However, if the main source of chemical species in the springs and groundwater of Arenal were the direct injection of volcanic gases to the hydrothermal system surrounding the magmatic chamber, a long-term decrease in HCO_3^- and partial pressure of CO_2 in the aquifer should not be observed. For that reason, the more likely source of chemical species at Arenal waters is leaching of hot volcanic products by infiltrating water. As water infiltration depends on rainfall, and rainfall has seasonal variations, that could explain the short-term variations in composition.

Volcanic belts around the world are highly populated areas, with many large cities located adjacent to dangerous volcanoes (e.g. Naples and Vesuvius volcano in Italy, Quito and Pichincha volcano in Ecuador). The scientific community has been trying to improve monitoring and prediction of volcanic eruptions, including efforts in understanding the hot spring chemistry at Etna volcano (Bonfanti et al., 1996), Vulcano Island (Capasso et al., 1999) and Popocatepetl Volcano (Martin-Del Pozo et al., 2002). Several authors have also studied the possible relationship between variations in water chemistry as precursors of seismic events (e.g. King et al., 1981; Toutain et al., 1997). Our investigations at Arenal Volcano suggest that the interpretation of variations in temperature, chemical composition, and seismic activity at volcanic systems requires careful consideration of other external factors such as the variations in Earth tides and atmospheric parameters. A change in chemical composition of spring discharges cannot be considered as precursory signal if the natural variabil-

ity of the data due to the external sources is not assessed. This requires a sampling frequency higher than the frequency of the expected variation. Two possible origins for variations in seismicity at volcanoes non-related to new magmatic inputs have been suggested: variations in Earth tide (e.g. Williams-Jones et al., 2001) or variations in atmospheric variables (Neuberg, 2000). However, which one of these two factors is dominant probably depends on the stage of the volcano activity. Volcanoes that are seismically active but are not in an eruptive stage and do not have an open conduit connected to the atmosphere probably behave differently from volcanoes, such as Arenal, whose magmatic chambers are openly discharging products to the atmosphere and are sensitive to changes in atmospheric pressure. In addition, climatic factors and their variability through time are probably very important for the water chemistry of volcanoes. Volcanoes such as Arenal (and in a wider spectrum, volcanoes in tropical areas), where precipitation is high (>5 m/year) and irregularly distributed through time, and cooling the hot eruptive products, probably present a different transient behavior in chemical composition of springs and groundwaters than active volcanoes located in dry climates such as those in Northern Chile.

At Arenal Volcano, seismic monitoring and water sampling during the frequent activity of the volcano has allowed us to observe trends that sporadic water sampling will never accomplish. The natural range of fluctuations in the data can be used for comparison and identification of anomalous values. However, due to the variability of atmospheric conditions, geological setting, state of activity, and volcanic type, more investigation is needed to find out if Earth tides or atmospheric conditions generate the changes in magma degassing and inputs to the hydrothermal systems observed at different volcanoes of the world, including Arenal. These studies include spectral analysis of the different parameters, such as Earth tides, number of seismic events, CO_2 , SO_2 and other plume gases, atmospheric pressure and temperature, rainfall and the composition of waters discharging at springs with a possible high connection with the magmatic environment.

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