ground water

A Mechanism for Anomalous Decline in Radon Precursory to an Earthquake

by M.C. Tom Kuo[1], K. Fan[2], H. Kuochen[3], and W. Chen[4]

Abstract

Mechanisms for interpreting anomalous decreases in radon in ground water prior to earthquakes are examined with the help of a case study to show that radon potentially is a sensitive tracer of strain changes in the crust preceding an earthquake. The 2003 Chengkung earthquake of magnitude (M) 6.8 on December 10, 2003, was the strongest earthquake near the Chengkung area in eastern Taiwan since 1951. The Antung radon-monitoring station was located 20 km from the epicenter. Approximately 65 d prior to the 2003 Chengkung earthquake, precursory changes in radon concentration in ground water were observed. Specifically, radon decreased from a background level of 780 pCi/L to a minimum of 330 pCi/L. The Antung hot spring is situated in a fractured block of tuffaceous sandstone surrounded by ductile mudstone. Given these geological conditions, we hypothesized that the dilation of brittle rock mass occurred at a rate faster than the recharge of pore water and gas saturation developed in newly created cracks preceding the earthquake. Radon partitioning into the gas phase may explain the anomalous decrease of radon precursory to the 2003 Chengkung earthquake. To support the hypothesis, vapor-liquid, two-phase radon-partitioning experiments were conducted at formation temperature (60°C) using formation brine from the Antung hot spring. Experimental data indicated that the decrease in radon required a gas saturation of 10% developed in rock cracks. The observed decline in radon can be correlated with the increase in gas saturation and then with the volumetric strain change for a given fracture porosity.

Introduction

Measurement of radon-222 in ground water has been previously investigated for use in earthquake prediction by a number of researchers (Igarashi et al. 1995; Liu et al. 1985; Noguchi and Wakita 1977; Roeloffs 1999; Silver and Wakita 1996; Teng 1980; Wakita et al. 1980). To assess long-term trends in radon concentrations in ground water, we began to study the Antung hot spring in eastern Taiwan ~3 km southeast of the Chihshang fault in July 2003 (Figure 1). The Chihshang fault is the most active segment of the Longitudinal Valley fault, which forms the present-day plate boundary between the Eurasian and Philippine Sea plates. The Chihshang fault (Hsu 1962) ruptured twice in 1951 during earthquakes of magnitude (M) 6.2 and (M) 7.0. The annual survey of geodetic and Global Positioning System (GPS) measurements consistently revealed active creeping of the Chihshang fault at a rapid steady rate of ~2 to 3 cm/year during the past 20 years (Angelier et al. 2000; Lee et al. 2003; Yu et al. 1990; Yu and Kuo 2001). A magnitude (M) 6.8 earthquake occurred at 4:38 am on December 10, 2003 (universal time), the strongest since 1951 near the Chengkung area in eastern Taiwan.

According to a worldwide survey (Hauksson 1981), most radon (Rn-222) anomalies associated with earthquakes show increases in radon concentration precursory to a rupture while few anomalies manifested decreases in radon. At the Antung hot spring (Kuo et al. 2006), radon decreased from a background level of 780 pCi/L to

[1]Corresponding author: Department of Mineral and Petroleum Engineering, National Cheng Kung University, Tainan, Taiwan; (886)-6-2757575 ext. 62827; fax (886)-6-2747378; mctkuobe@mail.ncku.edu.tw

[2]Department of Mineral and Petroleum Engineering, National Cheng Kung University, Tainan, Taiwan

[3]Central Weather Bureau, Taipei, Taiwan

[4]Department of Geosciences, National Taiwan University, Taipei, Taiwan

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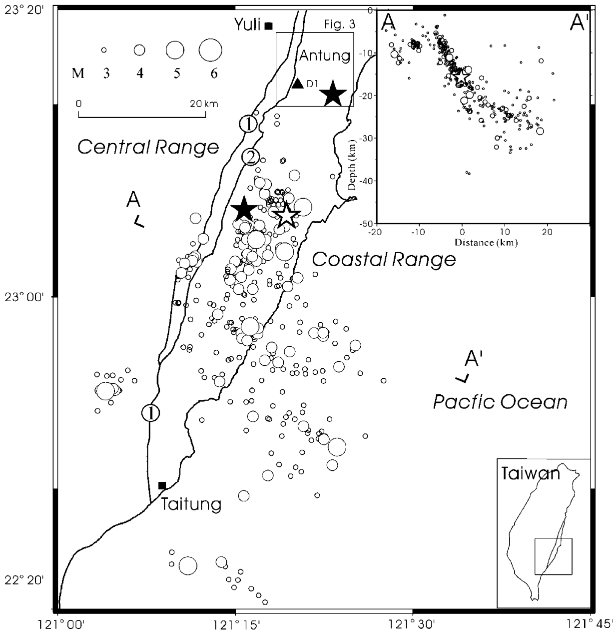

Figure 1. Map of the epicentral and hypocentral distributions of the mainshock and aftershocks of the 2003 Chengkung earthquake (open star: 2003 mainshock, open circles: 2003 aftershocks, filled stars: 1951 mainshocks, filled triangle: radon-monitoring well, ① Chihshang or Longitudinal Valley fault, ② Yongfeng fault) (adapted from Kuo et al. 2006).

a minimum of 330 pCi/L (Figure 2) prior to the 2003 Chengkung earthquake.

Radon-222 is a chemically inert radioactive nuclide with a half-life of ~3.8 d. The radon concentration in ground water is proportional to the uranium concentration in adjacent rocks. Transport processes of radon in geological environments include advection of radon dissolved in ground water, diffusion, partition between liquid and gas phases, and radioactive decay. Because of radon's short recoil length (3×10^{-8} cm), only atoms produced at the surface of rock grains are released to the surrounding ground water. Thus, the concentration of radon in ground water is largely dependent on the surface area of the rocks

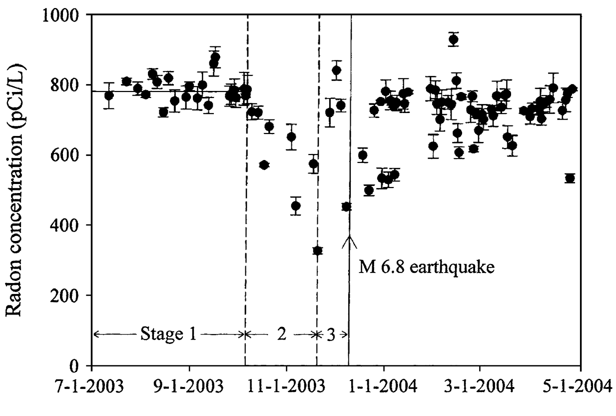

Figure 2. Radon concentration data at the monitoring well (D1) in the Antung hot spring. Stage 1 is buildup of elastic strain. Stage 2 is dilatancy and development of cracks and gas saturation. Stage 3 is influx of ground water and diminishment of gas saturation (adapted from Kuo et al. 2006).

(Torgersen et al. 1990). Before the occurrence of an earthquake, regional stress increases, causing formation of microcracks in the rock mass that could cause an increase in the surface area of the rocks. As a result, radon concentration increases (Igarashi et al. 1995; Teng 1980). However, mechanisms and geological conditions for interpreting anomalous decreases in radon prior to earthquakes are seldom discussed in the literature. The purpose of this paper is to investigate possible mechanisms to explain an anomalous decrease in the concentration of radon in ground water prior to an earthquake; data from the 2003 Chengkung earthquake (Kuo et al. 2006) are used in the analysis.

Both geological conditions near the Antung hot spring and the vapor-liquid phase behavior of radon were investigated to explore possible mechanisms causing the radon anomaly precursory to the 2003 Chengkung earthquake. The Antung hot spring is situated in a fractured block of tuffaceous sandstone surrounded by ductile mudstone. Based on experimental observations of rock dilatancy (Brace et al. 1966) and because the rocks in the Antung hot spring display intensively brittle deformation, we assumed that dilation of the rock mass occurred in the Antung hot spring precursory to the 2003 Chengkung earthquake. Under such geological conditions, we hypothesized that the dilation of the rock mass occurred at a rate faster than the recharge of pore water and gas saturation developed in newly created cracks preceding the earthquake. Radon partitioning into the gas phase may explain the radon anomaly precursory to the 2003 Chengkung earthquake. To support the hypothesis, vapor-liquid, two-phase radon-partitioning experiments were conducted for various levels of gas saturation at formation temperature (60°C) using formation brine from the Antung hot spring. Experimental data indicated that the anomalous decrease in radon concentration from 780 to 330 pCi/L required a gas saturation of 10% developed in the rock cracks. The 10% increase in gas saturation can be correlated with the volumetric strain change for a given fracture porosity. The proposed hypothesis is of practical interest because the observed decline in radon can be correlated with the increase in gas saturation and then with the volumetric strain change for a given fracture porosity.

Methods

Radon Determination

Radon was partitioned selectively into a mineral oil–based scintillation solution immiscible with the water sample (Noguchi 1964). After the sample was dark adapted and equilibrated, it was measured in a liquid scintillation counter using a region or window of the energy spectrum optimal for radon alpha particles. A calibration factor for the liquid scintillation counter measurements of 7.1 ± 0.1 counts per minute (cpm)/pCi was calculated using an aqueous Ra-226 calibration solution, which is in secular equilibrium with Rn-222 progeny. For a count time of 50 min and background <6 cpm, a detection limit below 18 pCi/L was achieved using the sample volume of 15 mL (Prichard et al. 1992).

Ground Water Sampling

A submersible pump was used for ground water sampling at a fairly constant flow rate of 110 L/min. Freyer et al. (1997) showed that radon concentration in ground water samples does not depend on the pumping rate. Prior to sampling, stagnant water was flushed from the well. Inadequate purging can be a major source of error because the water sample is a mixture of stagnant water from the wellbore and ground water influenced by the natural radon emanation rate of the aquifer. A minimum of five wellbore volumes was purged before taking samples for radon measurements.

It is important to ensure radon does not escape during sampling, sample transportation, and preparation. A 40-mL glass vial with a Teflon®-lined cap was used for collecting ground water samples of radon. After collecting a sample, the sample vial was inverted to check for air bubbles. If any bubbles were present, the sample was discarded and the sampling procedure repeated. The date and time of sampling were recorded and the sample stored in a cooler. The maximum holding time before analysis was 3.8 d for radon-222.

Radon Phase Behavior

To support the hypothesis of radon volatilization from ground water into the gas phase, radon-partitioning experiments were conducted to determine the variation of the radon concentration remaining in ground water with the gas saturation at formation temperature (60°C) using formation brine from the Antung hot spring. Five levels of gas saturation were investigated, specifically $S_g = 5\%$, 10%, 15%, 20%, and 25% where S_g is gas saturation. Triplicate experiments were conducted for each level of gas saturation. Every experiment started with 40 mL of formation brine. Five levels of headspace volume at 2, 4, 6, 8, and 10 mL were then created above the liquid phase for five levels of gas saturation at 5%, 10%, 15%, 20%, and 25%, respectively. Two-phase equilibrium was achieved for each experiment in 30 min at the formation temperature (60°C).

A kinetic study of radon volatilization from ground water into the gas phase was conducted to determine the time required to reach equilibrium. In the kinetic experiment, formation brine from the Antung hot spring with an initial radon concentration of 479 ± 35 pCi/L was used. Every sample started with 40-mL formation brine, and a headspace volume at 6 mL was then created above the liquid phase. A total of five samples were prepared. The radon concentration remaining in ground water was determined at various volatilization times (i.e., 2, 5, 15, 30, and 60 min). The time required to reach equilibrium for radon volatilization was only ~5 min.

Results

The observation well (D1, Figure 1) at the Antung hot spring, located ~20 km north of the hypocenter of the 2003 earthquake, was monitored for 10 months beginning in July 2003. Based on the distribution of the aftershocks, the faulting generated by the earthquake extended close to the observation well. The production interval of the well ranges from 167 to 187 m below ground surface and is pumped more or less continuously for water supply purposes. Discrete samples of geothermal water were collected for analysis of radon (Rn-222) twice per week. The liquid scintillation method was used to determine the activity concentration of radon-222 in ground water (Noguchi 1964; Prichard et al. 1992). The radon concentration was fairly stable (780 pCi/L in average) from July 2003 to September 2003 (Figure 2). Sixty-five days before the magnitude (M) 6.8 earthquake (December 10, 2003), the radon concentration of ground water started to decrease and continued to decrease for 45 d. Twenty days prior to the earthquake, the radon concentration reached a minimum value of 330 pCi/L and before starting to increase. Just before the earthquake, the radon concentration recovered to the previous background level of 780 pCi/L.

The main shock also produced a sharp anomalous coseismic decrease (~300 pCi/L). After the earthquake, some irregular variations were observed, which we interpret as an indication that the strain release by the main shock was not complete and that some accumulation and release of strain continued in the region.

Discussion

Environmental records such as temperature and rainfall were examined to check if the radon anomaly was correlated with these parameters. Ground water temperature was very stable during the observation period. There was no heavy rainfall during the period. Moreover, it would be difficult to explain such a large radon decrease by mixing effects.

The hypocentral distributions of aftershocks of the 2003 Chengkung earthquake show the close relation between seismicity and the Chihshang fault, which has a faulting surface extending ~30 km in depth and dips ~50° to the southeast (Figure 1). The focal mechanism of the mainshock was a thrust with strike of N36°E and dip of 50°SE. The radon-monitoring well (D1) in the Antung hot spring is ~3 km southeast of the Chihshang fault. The anomalous decrease in radon concentration observed at the Antung hot spring suggests that concentration of radon in ground water can be a sensitive tracer for strain changes in crust associated with earthquake occurrences (Trique et al. 1999; Roeloffs 1999; Silver and Wakita 1996). A detailed investigation of nearby geological conditions would help understand the physical basis causing the radon anomaly and select sites for future monitoring.

The studied region (Figure 3) is in a unique tectonic setting located at the boundary between the Eurasian and Philippine Sea plates near the Coastal Range. Four stratigraphic units are present. The Tuluanshan Formation consists of volcanic units such as lava and volcanic breccia as well as tuffaceous sandstone. The Fanshuliao and Paliwan Formations consist of rhythmic sandstone and mudstone turbidites. The Lichi mélange occurs as a highly deformed mudstone that is characterized by penetrative foliation visible in outcrop. The Antung hot spring is situated in a brittle tuffaceous sandstone block surrounded by a ductile mudstone of the Paliwan Formation (Chen and Wang 1996). Well-developed minor faults and joints are

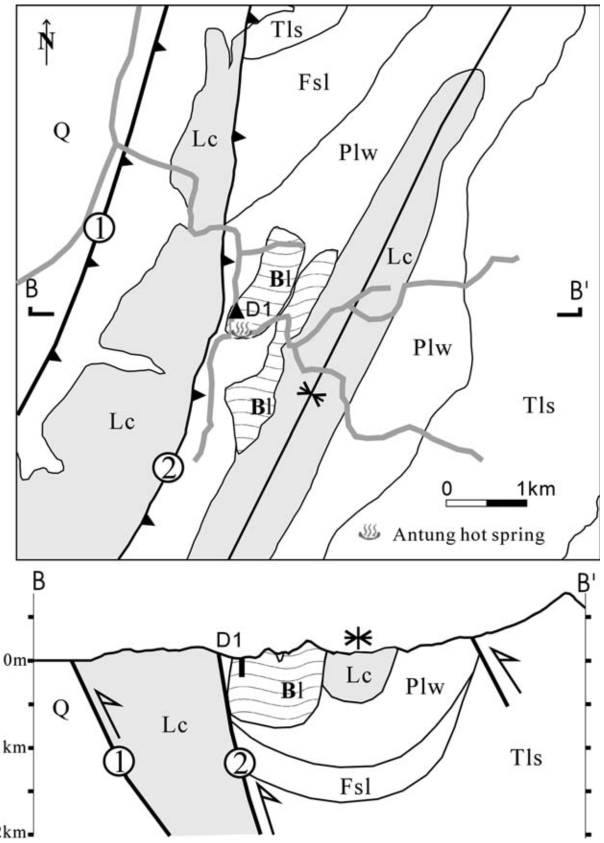

Figure 3. Geological map and cross section near the radon-monitoring well in the area of Antung hot spring (Q: Holocene deposits, Lc: Lichi mélange, Plw: Paliwan Formation, Fsl: Fanshuliao Formation, Tls: Tuluanshan Formation, Bl: tuffaceous fault block, D1: radon-monitoring well, ① Chihshang or Longitudinal Valley fault, ② Yongfeng fault). See Figure 1 for map location.

common in the tuffaceous sandstone block displaying intensively brittle deformation. It is possible that these fractures reflect deformation and disruption by the nearby faults. Hence, geological evidence suggests the tuffaceous sandstone block displays intensively brittle deformation and develops in a ductile deformed mudstone stratum. Ground water flows through the fault zone and is then diffused into the block along the minor fractures.

Wu et al. (2005) investigated the dislocation fault model of the 2003 Chengkung earthquake using a computer code by Okada (1992). Based on the fault geometry defined by the aftershock distribution and geology (Central Geological Survey, Taiwan 2000a, 2000b), Wu et al. (2005) assumed a thrust fault with strike N20°E, parallel to the Coastal Range (Figure 1), with a bend at a depth of 18 km. The fault plane dips 60°SE and 45°SE above and below 18 km, respectively (see cross section AA′ in Figure 1). The lower and upper ruptured fault planes are abbreviated as fault A and fault B, respectively. For the best fit with the coseismic ground deformation both fault planes extend 33 km at most from north to south. Faults A and B ruptured within depths of 18 to 36 km and 5 to 18 km, respectively. Fault A slipped 61.6 cm with a rake of 81.7°, while fault B slipped 26 cm with a rake of 47.3°. Based on the area and slip on the ruptured surface of each fault, we found Mw (moment magnitude scale) 6.7 and Mw 6.3 for fault A and B, respectively. The total Mw was ~6.8 and agreed with the result of the moment tensor inversion solution from the Harvard CMT database (http://www.seismology.harvard.edu/), indicating that the coseismic energy was mainly released by fault A. We calculated the coseismic strain distribution due to the 2003 Chengkung earthquake based on the dislocation fault model (Wu et al. 2005). The calculated contractional surface strain near the Antung hot spring area was ~20 parts per million (ppm) (Figure 4).

While the Antung hot spring experienced compression, the properties of the rocks (Figure 3) have direct implications on whether the rocks undergo dilation or contraction. The assumption of linear rock elasticity is valid for the Lichi mélange and Paliwan and Fanshulia formations, which are predominantly mudstone (Chen and Wang 1996). However, the Antung hot spring is situated in a tuffaceous sandstone block (Figure 3) displaying intensively brittle deformation. Brace et al. (1966) investigated stress–volumetric strain behavior for granitic rocks with crack porosity during deformation in triaxial compression. According to the experimental observations of Brace et al. (1966), volume changes are purely elastic at low stress; as the maximum stress becomes one-third to two-thirds the fracture stress, the rocks become dilatant. Dilatancy, which is accompanied by an increase in porosity, was traced in the granite to open cracks that form

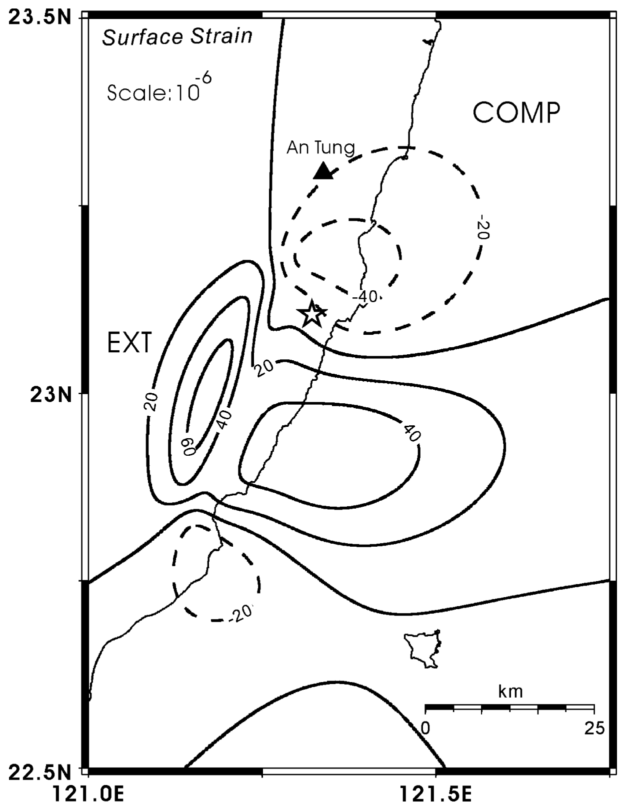

Figure 4. Distribution of coseismic surface strain (ppm) calculated based on the computer code for dislocation models by Okada (1992). Positive and negative values mean dilatation and contraction, respectively. The open star denotes the 2003 mainshock. The filled triangle denotes the radon-monitoring well (D1). EXT and COMP denote dilatation and contraction, respectively.

parallel with the direction of maximum compression. Based on experimental observations of rock dilatancy (Brace et al. 1966) and given that the rocks in the Antung hot spring display intensively brittle deformation, we assumed that there was dilation of the rock mass in the vicinity of the Antung hot spring precursory to the 2003 Chengkung earthquake.

Under geological conditions such as those of the Antung hot spring, we hypothesized that when regional stress increases, dilation of the rock mass occurs at a rate faster than the rate at which pore water can flow into the newly created pore volume (Brace et al. 1966; Scholz et al. 1973). During this stage (stage 2 in Figure 2), gas saturation and two phases (vapor and liquid) develop in the rock cracks. Meanwhile, the radon in ground water volatilizes and partitions into the gas phase and the concentration of radon in ground water decreases. Thus, the sequence of events for radon data prior to the 2003 Chengkung earthquake (Figure 2) can be interpreted in three stages. From July 2003 to September 2003 (stage 1), radon was fairly stable (~780 pCi/L). During this time, there was an accumulation of tectonic strain, which produced a slow, steady increase of effective stress. Sixty-five days before the magnitude (M) 6.8 earthquake, the concentration of radon started to decrease and reached a minimum value of 330 pCi/L 20 d before the earthquake. During this 45-d period (stage 2), dilation of the rock mass occurred and gas saturation developed in cracks in the rock and radon volatilized into the gas phase. Stage 3 started at the point of minimum radon concentration when water saturation in cracks and pores began to increase and radon increased and recovered to the background level. The main shock produced a sharp coseismic anomalous decrease (~300 pCi/L). After the earthquake, some irregular variations were observed, which we attribute to strain release as some accumulation and release of strain continued in the region.

Data from the vapor-liquid, two-phase equilibrium radon-partitioning experiments (Figure 5) were regressed with the two-phase partitioning model to determine Henry's coefficient as follows.

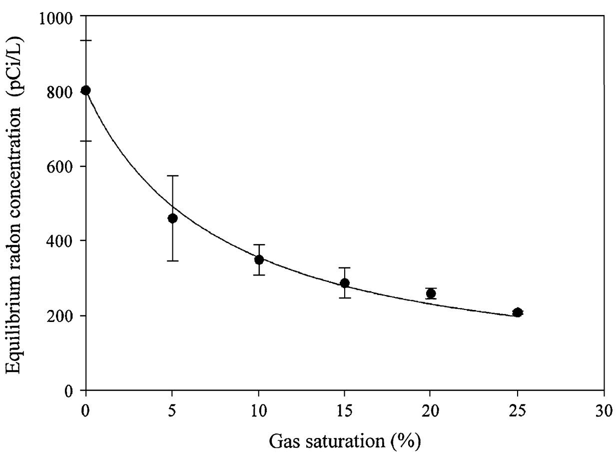

Figure 5. Variation of radon concentration remaining in ground water with gas saturation at 60°C using formation brine from the Antung hot spring.

$$C_0 = C_w(H \times S_g + 1)$$

where C_0 is initial radon concentration in the formation brine, pCi/L; C_w is equilibrium radon concentration in ground water, pCi/L; S_g is gas saturation, %; H is Henry's coefficient for radon, dimensionless. Figure 5 shows the regressed line with $H = 12.8$ and $R^2 = 0.9919$ (regression coefficient). Henry's coefficient for radon at 60°C determined for the Antung formation brine (12.8) is higher than the value (7.58) for water at 60°C. Radon solubility is markedly reduced when electrolytes are present (Rogers 1958). Figure 5 can be used to estimate the amount of gas saturation required for various decreases in concentration of radon. For example, the anomalous decrease of radon concentration from 780 to 330 pCi/L required a gas saturation of 10% in cracks in the rock.

It is of practical interest to correlate the 10% increase in gas saturation with the volumetric strain change for a given fracture porosity. According to fracture porosity and permeability data for naturally fractured rocks (Snow 1968), the average fracture porosity for all data (ranging from 0.00008 to 0.0003) is 0.00011. Assuming an average fracture porosity of 0.00011 for the Antung hot spring, we calculated a volumetric strain change of 11.0 ppm for the 10% increase in gas saturation due to rock dilation. Given a fracture porosity of 0.00008 and 0.0003, we calculated a volumetric strain change of 8.0 and 30.0 ppm, respectively, for the 10% increase in gas saturation due to rock dilation. It is recommended in future work to confirm the increase in gas saturation by in situ methods, such as strain, formation pressure, and neutron data.

Conclusions

1. A decline in radon concentration can be correlated with an increase in gas saturation and then with the volumetric strain change for a given fracture porosity.
2. Radon partitioning into the gas phase may explain the anomalous decrease in radon concentrations in ground water precursory to the 2003 Chengkung earthquake.
3. Experimental data indicated that the anomalous decrease of radon concentration from 780 to 330 pCi/L prior to the 2003 Chengkung earthquake required a gas saturation of 10% developed in cracks in the rock.
4. Assuming an average fracture porosity of 0.00011, a volumetric strain change of 11.0 ppm could be calculated for the 10% increase in gas saturation.
5. Observations at the Antung hot spring suggest that radon concentrations in ground water, under suitable geological conditions, can be a sensitive tracer of strain changes in the crust preceding an earthquake.

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