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Exhalation of radon and its carrier gases in SW Taiwan

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Abstract

Gas compositions of mud volcanoes reveal multiple sources for gas exhalation in SW Taiwan. For comparison, two sites, Yan-chao (YC) and Chung-lun (CL), were chosen for measurements of soil Rn concentrations using a portable radon detector. The ^{222}Rn concentrations at the YC site were ca. 5200 Bq/m^3. However, the average ^{222}Rn concentrations at the CL site exhibited higher value of ca. 16, 800 Bq/m^3. With the reference of the gas flux and compositions from the nearby mud pool, the soil ^{222}Rn concentrations are largely controlled by the flux of carrier gases exhaled from deep reservoirs.

Keywords: Radon exhalation; Carrier gases; Mud volcano; ^{3}He/^{4}He ratio; Taiwan

1. Introduction

Variations of radon concentrations in the soil and fluid samples are considered a useful tool for earthquake monitoring and prediction in active fault zones (Chyi et al., 2001; Igarashi et al., 1995; King et al., 1996; Toutain and Baubron, 1999; Virk et al., 2001), for surveillance in volcanic areas (Heiligmann et al., 1997; Segovia et al., 2001; Varley and Armienta, 1999) and for tracing neotectonic faults (Ciotoli et al., 1999; Etiope and Lombardi, 1995; Guerra and Lombardi, 2001). Recently, Etiope and Martinelli (2002) suggested that carrier gases (CO_2 and CH_4) may play a dominant role in controlling transport and redistribution of trace gases (Rn and He) towards the Earth's surface. Thus, studies on the distribution and behavior of trace gases, such as radon, near the Earth's surface may not be meaningful unless accompanied by analysis of carrier gas dynamics.

Mud volcanoes are widely distributed along the tectonic sutures in SW Taiwan. Some are located in the active fault zones. Radon is continuously exhaling from these mud volcanoes accompanied with other major gases, for examples, CH_4 and CO_2 (Chou, 2002). To better understand the factors controlling radon exhalation, two sites, Yan-chau (YC) and Chung-lun (CL), in this area were chosen for soil radon analysis (Fig. 1). Combined with other available gas composition data and flow flux, we will propose exhalation models to explain radon concentrations in SW Taiwan.

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2. Multiple gas sources in SW Taiwan

Gases exhaled from mud volcanoes in SW Taiwan are dominantly CH_4 with low helium isotopic ratios (^{3}He/^{4}He $< 0.2R_a$, where R_a is the air ratio), which indicates a crustal source in origin (Yang, 2002). YC is a typical mud volcano located at the suspected active fault zone in the area (Fig. 1), emanating CH_4-dominated gases which contain only trace amounts of radon (Table 1). Nevertheless, bubbling gases from a mud pool in CL, which is situated in the Chu-ko (CK) active fault zone with a potential for re-activation in the future (Fig. 1), show much higher helium isotopic ratios (^{3}He/^{4}He $=$ 5.2–6.6R_a), CO_2-dominated compositions and much higher radon concentrations (Table 1). This implies that there are multiple sources for gas exhalation in SW Taiwan. Yang et al. (2003) suggested that variations in gas composition are very sensitive to tectonic stress and/or strain and hence may be suitable for further earthquakes monitoring.

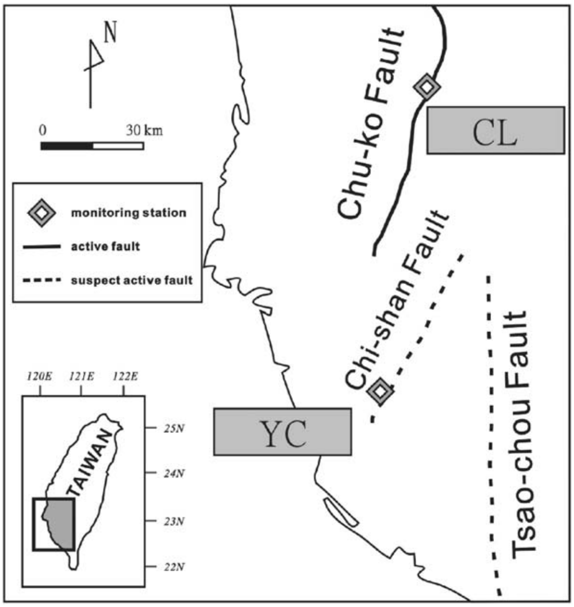

Fig. 1. Locations of automated Rn monitoring stations in south-western Taiwan. Yan-chau (YC) station is located on a suspect active fault. Chung-lun (CL) station, however, is located at the fault zone of Chu-ko active fault.

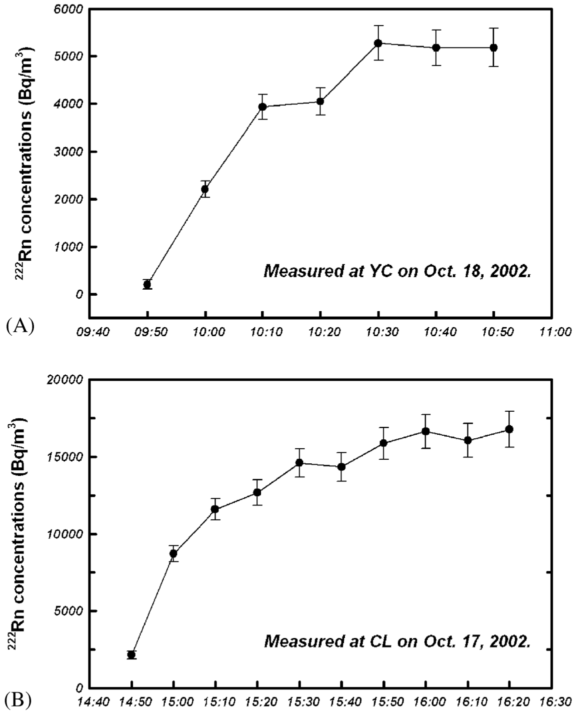

Fig. 2. Soil radon concentrations were measured in situ using an AlphaGuard® PRO 2000 portable radon detector: (A) YC station; and (B) CL station. Note different scales used for the figures. Radon concentrations at CL station were much higher than those in YC station.

3. Results and discussion

3.1. Soil radon concentrations and exhalation rates

Owing to the significant differences in gas compositions and sources between the YC and CL sites, these two sites were chosen for continuously monitoring of soil radon variations using an automated monitoring system equipped with a silicon photodiode detector (Chou, 2002; Chyi et al., 2001). By enclosing a radon detector and a data logger inside a polyvinyl chloride (PVC) pipe, packing the pipe with polymer pellets and then sealing the system with polyurethane foam, the environment factors which affect radon concentration could either be reduced or eliminated.

Table 1
Compositions[a] of representative bubble gases from mud volcanoes at Yan-chao (YC) and Chung-lun (CL), SW Taiwan

Sample no. (date—xx)	CH_4 (%)	N_2 (%)	O_2 (%)	Ar (%)	CO_2 (%)	$^3He/^4He$ ($\times 10^{-6}$)	$[Ra]_c$ [b]	$[^4He]$ (ppm)	^{222}Rn (Bq/m^3)
00925-YC	—	—	—	—	—	0.299	0.20 ± 0.01	26.7	—
01007-YC	93.02	4.69	1.24	0.076	0.98	—	—	—	36 ± 13
01011-YC	97.63	1.14	0.23	0.019	0.97	—	—	—	59 ± 23
01016-YC	—	—	—	—	—	0.315	0.21 ± 0.01	21.6	—
00810-CL	7.76	8.06	1.40	0.088	82.70	8.531	6.17 ± 0.15	55.0	7100 ± 1700
00930-CL	12.10	10.62	2.34	0.170	74.77	7.133	5.28 ± 0.13	13.1	9200 ± 900
01007-CL	4.54	12.28	2.33	0.130	80.73	8.446	6.20 ± 0.15	29.2	4100 ± 250
01022-CL	14.25	10.76	0.89	0.055	74.05	7.970	5.77 ± 0.14	40.3	3600 ± 200

[a] Major compositions were carried out using a quadrupole mass spectrometry; typical analytical error is less than 5%. Helium concentrations and isotopic ratios were analyzed with a Micromass® 5400 noble gas mass spectrometer, with air as a routine normalization standard; the typical errors are about 5% and 2% for He concentration and isotopic ratio, respectively. Radon concentrations were measured in situ using an AlphaGUARD® PRO 2000 radon detector with an internal pump.

[b] Ra is the $^3He/^4He$ ratio of air (1.39×10^{-6}), $[Ra]_c$ is the air corrected ratio, assuming all the neon in the sample come from air.

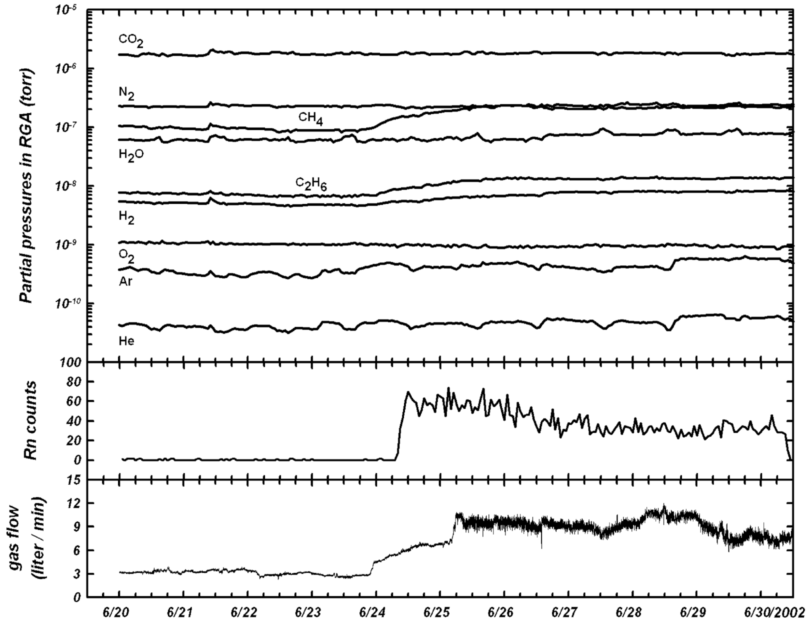

Fig. 3. Variations of flux and compositions of bubbling gases from mud pool at the CL site. Radon concentrations are strongly controlled by the flux rate.

Table 2
Activity concentrations for radioactive isotopes in representative soil samples

Sample no.	Sample weight (kg)	^{40}K (Bq/kg)	^{232}Th (Bq/kg)	^{238}U (Bq/kg)	Remarks[a]
YC-1	0.181	526 ± 3%	29 ± 6%	17 ± 6%	YC station
YC-2	0.173	533 ± 3%	28 ± 6%	18 ± 6%	YC mud pool
YC-3	0.198	559 ± 3%	33 ± 7%	19 ± 6%	YC mud pond
CL-1	0.158	561 ± 3%	35 ± 5%	16 ± 7%	CL station
CL-2	0.223	477 ± 3%	34 ± 5%	18 ± 6%	CL mud pool

[a]Soil samples from the monitoring stations were collected at a depth of 10 cm; samples from mud pool/pond were collected at the location of gas bubbling.

Chyi et al. (2001, 2002) recognized the spike-like anomalous peaks usually lasting a few minutes to a few hours, with some lasting a few days, and always occurring a few days before an earthquake. Hence, they concluded that these concentrations are sensitive to earthquakes and may be useful for further earthquake monitoring and prediction. However, a discussion of the relationship between anomalous peaks and earthquakes is beyond the scope of the present study, which will focus only on radon background exhalation.

To further check absolute radon concentrations exhaled from soil at both stations, these concentrations were measured in situ using an AlphaGUARD® PRO 2000 portable radon detector at a pumping speed of 0.3 l/min. Fig. 2 shows radon concentrations reaching an equilibrium value of ca. 5200 and 16,800 Bq/m^3 in 1 h for the YC and CL sites, respectively. This not only demonstrates differences in radon levels at the two sites, but also demonstrates that the radon exhalation rate of ca. 17,000 Bq/m^3 per hour measured at CL is much higher than the ca. 5000 Bq/m^3 per hour at YC.

3.2. Flux and composition variations of the bubble gases

A multi-parameter gas monitoring station was set up at the CL mud pool and gases collected through the water replacement method using an up-side-down funnel. Gases collected were continuously transferred into the analyzing system. The main analysis system was equipped with a quadruple mass spectrometer and a radon detector. This setup enabled measurement of gas compositions (including H_2, He, CH_4, H_2O, N_2, O_2, Ar, CO_2, C_2H_6 and Rn) and gas flow of bubbling gases from the mud pool at the CL site at intervals of less than 2 min. Details of setup, analytical procedures and errors are discussed in Yang et al. (2003).

Selective monitoring results are shown in Fig. 3. Interestingly, a significant increase of gas flow, from 3 to 10 l/min, was observed on June 24, 2002. Associated gas compositions, including Rn and CH_4, also show significant variations. This implies that variations in radon concentrations in this area are mainly controlled by the flux of the carrier gases.

4. Radon exhalation models

Radon exhaled both from soil samples and bubbling gases at the YC and CL sites reveal distinct concentration levels for each site (Table 1 and Fig. 2). Recent experimental results suggested that the combination of grain size and uranium content has an impact on the emanation of radon gas from the soil (Baixeras et al., 2001). Hence, different radon emanation rates in soils may be a major factor to control for checking radon levels in these two sites. To confirm this point, representative clay samples with similar grain sizes from the YC and CL sites were used for measurement of activity concentrations of radioactive isotopes using gamma-ray spectrometry (Table 2). All samples exhibited similar concentrations; hence, emanation of radon from the soil samples themselves cannot account for the difference of radon concentrations at these two sites.

In contrast to earlier views, Etiope and Martinelli (2002) argued that the role of gas diffusion and water advection in the transport of gases to the surface should be minimized and suggested that carrier gases, e.g., CH_4 and CO_2, may play an important role in controlling the migration and transportation of trace gases such as Rn and He towards the surface. This argument was supported by previous experimental result (Varhegyi et al., 1992). Hence, they concluded that bubble movement in fissured rocks is an effective means of rapid, long-distance gas migration.

The CL site is located in a complicated-fracture zone where several faults have merged together (Chyi et al., 2001). It is thus able to provide a pathway for outgassing of deep source gases towards the surface. Indeed, gas flux strongly correlates with radon concentrations in the bubble gases in the CL mud pool (Fig. 3), which could explain the higher background radon concentration and exhalation rate observed at CL compared with those in YC, where the bedrock may not be as complicated as at CL.

Fig. 4 shows proposed models for radon exhalation in southwestern Taiwan. Radon exhalation is mainly controlled by the CH_4 carrier gases in the YC area (Fig. 4A). The relatively low carrier gas exhalation rate may itself explain the very low radon background concentrations. However, in case of tectonic stress and strain, CH_4 may be suddenly expelled from deep reservoirs and produce anomalous radon peaks (Chou, 2002).

In contrast to the simple gas sources observed at YC, gases from CL area exhibit much more complex gas source

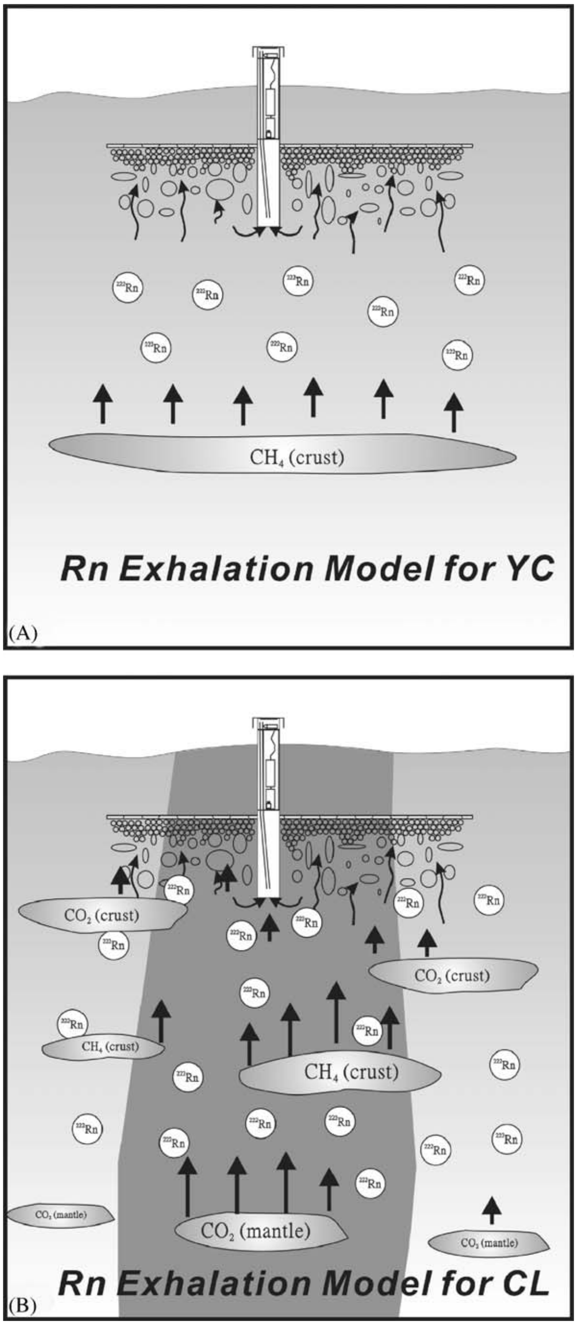

Fig. 4. Radon exhalation models for YC and CL sites in SW Taiwan. After emanated from soil and rocks, radon atoms are transferred to the surface by carrier gases. (A) At YC station, CH_4 is dominated and likely to be the carrier gas for radon. However, (B) the CL area has more complex gas sources, hence multiple carrier gases from different depths rising via the fractured zone (shaded area) are necessary to explain radon concentrations.

components. At least three gas source components can be identified in this area: (1) a magmatic CO_2 component with high $^3He/^4He$ ratios; (2) a crustal CH_4 component with low $^3He/^4He$ compositions; (3) a crustal CO_2 component with low $^3He/^4He$ compositions (Yang, 2002). Accordingly, different gas source reservoirs may reside at different depths, and migrate to the surface, carrying radon, along the deep fractured zones of the CK fault (Fig. 4B).

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