Radon distribution in groundwater of Taiwan

**Y. L. Han · M. C. Tom Kuo · K. C. Fan · C. J. Chiang ·
Y. P. Lee**

Abstract Radon levels were surveyed in 517 monitoring wells constructed in five major groundwater areas of Taiwan. The radon concentration in groundwater samples varied in a wide range from below the detection limit of 18 pCi/L up to 1,100 pCi/L. A worldwide comparison of reported groundwater radon levels was conducted. Overall radon levels in Taiwan groundwater are relatively low compared to other countries because the geology of Taiwan is mainly comprised of sedimentary rocks. Among the 517 wells monitored, only five wells were found with radon concentrations higher than 500 pCi/L. These five wells are all located near the Chaochou Fault in the Pingtung Plain. This study suggests that well sites near the Chaochou Fault could be good locations to monitor radon anomalies for earthquake prediction and should be avoided for developing domestic water supply. In the recharge area near the Chaochou Fault, the radon concentration in groundwater from shallow wells was approximately 1/2 to 1/4 of that from deep wells in the same cluster.

Resumen Se investigaron los niveles de radón en 517 pozos de monitoreo, construidos en las cinco mayores áreas de agua subterránea de Taiwan. La concentración de radón en las muestras de agua subterránea varía en un rango amplio, desde inferior al límite de detección que es 18 pCi/L hasta 1,100 pCi/L. Se realizó una comparación a escala mundial de los niveles de radón reportados en agua subterránea. En general los niveles de radón presente en el agua subterránea de Taiwan, son relativamente bajos comparados con otros países, puesto que la geología de Taiwan está compuesta de rocas sedimentarias. Entre los 517 pozos monitoreados, solamente en cinco se encontraron concentraciones de radón mayores a 500 pCi/L. Estos cinco pozos están todos localizados cerca de la falla ChaoChou en la Planicie de PingTung. El presente estudio sugiere que los pozos cercanos a la falla Chaochou, podrían ser buenos sitios para monitorear anomalías de radón para la predicción de terremotos y deberían evitarse para los desarrollos de abastecimiento de agua potable. En la zona de recarga cerca de la falla Chaochou, la concentración de radón en agua subterránea obtenida en pozos someros, fue aproximadamente de ½ a ¼ de aquella en pozos profundos ubicados en el mismo sector.

Résumé On a mesuré les taux de Rn dans 517 forages ouverts dans cinq aquifères majeures de Taiwan. Les concentrations en Rn couvrent un domaine très large, à partir de valeurs très basses, au dessous de la limite de détection de 18pCi/l jusqu'aux valeurs assez grandes de 1100pCi/l. On a mené une analyse comparative avec des valeurs mentionnées en littérature. Par report aux valeurs mesurées en autres pays, les concentrations en Rn en Taiwan sont asses basses, compte que la structure géologique de Taiwan est constituée surtout par des roches sedimentaires. On a trouvé des concentrations au desuss de 500pCi/l seulement en cinq de 517 forages investigués. Tous ces cinq forages se trouvent au voisinage de la faille de Chaochou dans la plaine de Pingtung. Cet étude suggere que les forages situés près de la faille de Chaochou peuvent bien monitoriser les anomalie de Rn pour la prédiction des séismes mais doivent être évités lorsq'il s'agit de l'alimentation en eau potable. Dans la zone de recharge près de la faille de Chaochou les concentrations en Rn mesurées dans les puits peu profondes representent 1/2-1/4 de ceux mesurées dans les forages de profondeur.

Keywords Radon-222 · Fault · Groundwater recharge

Received: 2 February 2004 / Accepted: 2 August 2004

Y. L. Han · M. C. Tom Kuo (✉) · K. C. Fan
Department of Mineral and Petroleum Engineering,
National Cheng Kung University,
Tainan, Taiwan
e-mail: mctkuobe@mail.ncku.edu.tw

C. J. Chiang
Central Geological Survey,
Ministry of Economic Affairs,
Taipei, Taiwan

Y. P. Lee
Water Resources Agency,
Ministry of Economic Affairs,
Taipei, Taiwan

DOI 10.1007/s10040-004-0384-7

Introduction

During past years, public attention and many discussions were focused on radon-222, a naturally occurring, radioactive noble gas that is present in the environment in water, air and soil. UNSCEAR (1988) reported that the contribution to the natural radiation exposure of the general public from inhalation of radon-222 and its short-lived decay products is estimated to be about 50% (1.1 mSv year^{-1}). The average annual exposure of the population in Taiwan to natural radiation amounts to 1.56 mSv. About 0.36 mSv is caused by radon-222 and its daughter products. An important part of this dose can be caused by groundwater, which is used for drinking water and spa purposes. There is no systematic investigation conducted to survey radon levels in Taiwan groundwater. The purpose of this study was to survey radon distribution in groundwater in five major hydrologic areas of Taiwan. Results of this survey were compared with results of other surveys conducted worldwide.

Measurement of radon-222 in groundwater has also been performed for earthquake prediction (Noguchi and Wakita 1977; King 1978; Liu et al. 1986; Igarashi et al. 1995). Igarashi reported precursory changes in radon concentration in groundwater associated with the 1995 Kobe earthquake (Igarashi et al. 1995). The other purpose of the present survey was to collect background data on groundwater radon distribution and to select well locations for planning a radon monitoring system to predict earthquakes.

Materials and Methods

Study area

Figure 1 shows the name, location, and extent of nine major groundwater areas in Taiwan. Radon levels were surveyed in five of the areas, i.e., Choshui River Alluvial Fan, Tainan Plain, Pingtung Plain, Ilan Plain and Hsinchu-Miaoli Coastal Area. The number of monitoring wells sampled in each area was 175, 142, 128, 39, and 33 for Choshui River Alluvial Fan, Tainan Plain, Pingtung Plain, Ilan Plain and Hsinchu-Miaoli Coastal Area, respectively.

Yearly total groundwater consumption in Taiwan is about 7,100 × 10^6 m^3 distributed as shown in Fig. 1 for each major groundwater area in Taiwan. Yearly groundwater consumption in the five studied areas combined is around 6,300 × 10^6 m^3, which represents about 89% of the total groundwater consumption in Taiwan.

Sample Collection

Accurate sampling for radon measurements depends on appropriate monitoring wells. Because radon concentration in groundwater relates to emanation rates of geological layers, representative sampling must be from properly constructed wells which enable vertically spaced water samples to be taken and contain only a limited quantity of filter pack and a suitable type of bentonite seal.

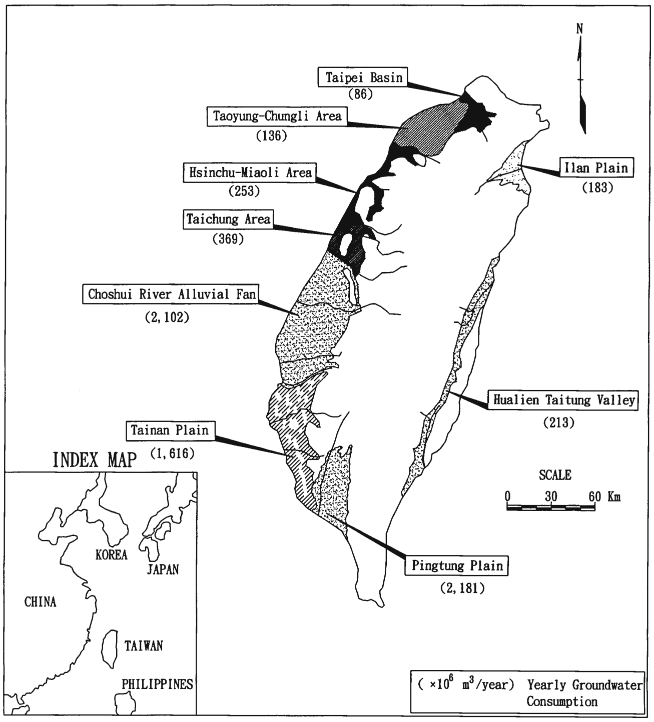


Fig. 1 Major groundwater areas in Taiwan and yearly groundwater consumption

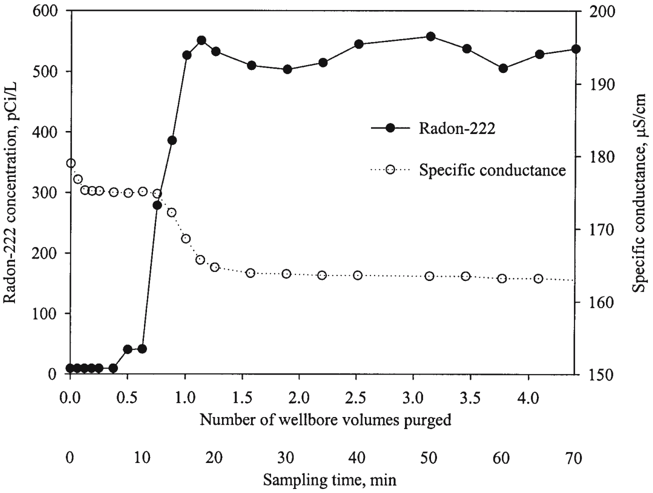


Fig. 2 Radon concentration and electrical conductivity in the well discharge during continuous sampling in a deep observation well

A submersible pump was used in monitoring wells for continuous sampling. Every sampling exercise starts with the flushing of stagnant water in the well and especially in the screen zone. Inadequate purging can be a major source of error, because the water sample is a mixture of stagnant water from the well bore, pore water from the filter gravel and groundwater influenced by the natural emanation rate of the aquifer. Figure 2 shows the radon concentration in

DOI 10.1007/s10040-004-0384-7

the well discharge during continuous sampling in a monitoring well. During the first period of flushing, the radon concentration of the water samples is practically zero and then increases rapidly to 529 pCi/L. The mean radon concentration measured for this monitoring well was 529 ± 19 pCi/L (11 samples). A minimum of three well-bore volumes was purged before taking samples for radon measurements.

A 40-ml glass vial with a TEFLON lined cap was used for sample collection. 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 was recorded and the sample stored in a cooler. The maximum holding time before analysis was four days.

Radon Determination

For the determination of the activity concentration of radon-222 in groundwater, a modified method described by Prichard and Gesell (1977) was adopted. Radon was partitioned selectively into a mineral-oil scintillation cocktail immiscible with the water sample (Noguchi 1964). The sample was dark-adapted and equilibrated, and then counted in a liquid scintillation counter (LSC) using a region or window of the energy spectrum optimal for radon alpha particles (Lowry 1991).

Radon concentrations were determined by drawing a 15-ml sample directly from a field sample into a clean syringe. Care was taken to prevent aeration of the samples in the process. The samples were then injected beneath a 5-ml layer of mineral-oil-based scintillation solution in 24-ml vials. The vials were vigorously shaken to promote phase contact, dark-adapted and held for at least three hours to ensure equilibrium between radon-222 and its daughters, and then assayed with a liquid scintillation counter. The results were corrected for the amount of radon decay between sampling and assay.

The results of the measurements were determined in units of counts per minute (cpm). It was essential to ensure that only the activity of radon-222 was measured. Using the TRI-CARB software of Packard 1600TR, it was possible to view the alpha spectrum (Fig. 3). The peaks of radon-222 (5.49 MeV), polonium-218 (6.00 MeV) and polonium-214 (7.69 MeV) can be distinguished.

A calibration factor for the LSC measurements of 7.1 ± 0.1 cpm/pCi (Fig. 4) 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 less than 6 cpm, a detection limit below 18 pCi/L was achieved using the sample volume of 15 ml (Prichard et al. 1992).

Verification of radon-222 as the radioisotope responsible for activity in the well water tested was obtained by the repeated counting of three samples from two wells. The half-life of 3.841 days experimentally determined for samples from Well Liu-Ying (I) located in Tainan Plain compares favorably with the accepted value of 3.825 days as shown in Fig. 5. If the counting vials lack tightness,

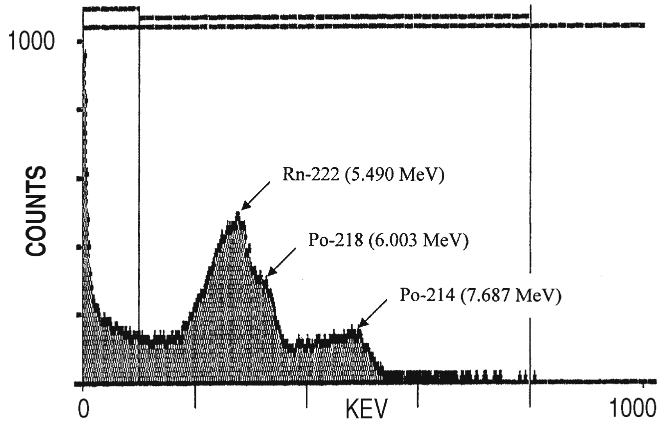


Fig. 3 Alpha spectrum of radon-222 and its daughter nuclides represented by TRI-CARB software

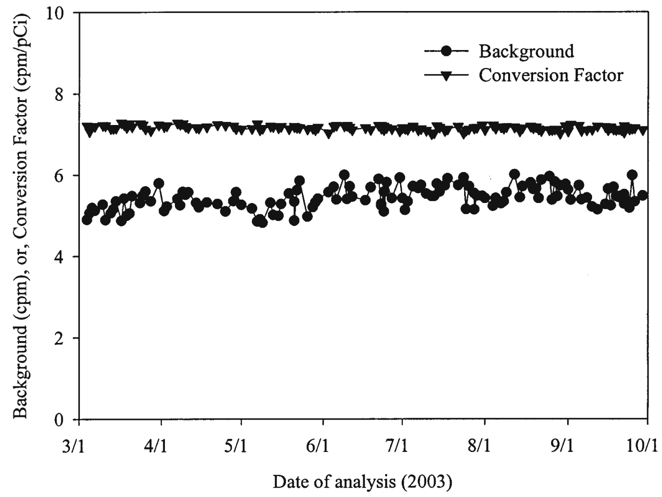


Fig. 4 Calibration factor and background for LSC measurements

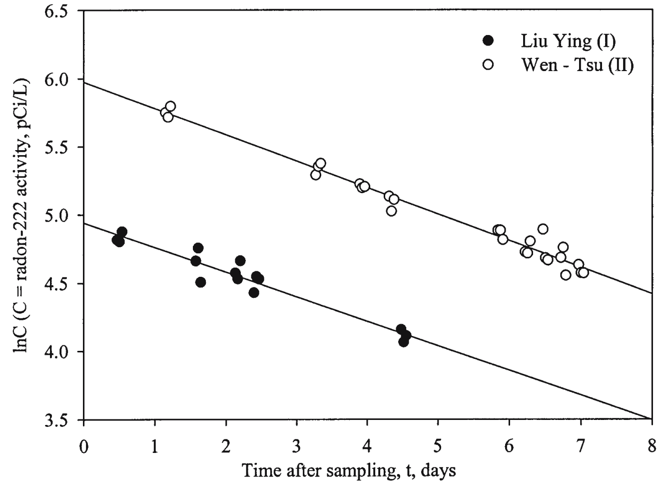


Fig. 5 Measurement of half life from semi-logarithmic decay curve

DOI 10.1007/s10040-004-0384-7

radon will escape from the counting vials and the half-life times experimentally determined for samples will be apparently shorter. Figure 5 also shows an example of such a case from Well Wen-Tsu (II) located in Choshui River Alluvial Fan.

Results and Discussion

Radon Levels in Groundwater of Taiwan

A total of 517 wells were sampled from five groundwater areas of Taiwan. Figure 6 is a histogram of combined radon-222 concentrations in five of the groundwater areas. All distributions in the five areas individually are also highly skewed (flat to the right) similar to Fig. 6. The combined radon-222 concentration in the five areas roughly followed a log-normal distribution that usually occurs for all natural elements. Figure 7 further shows plots of radon-222 concentrations versus cumulative frequency for the five areas individually and combined. As the data are close to a straight line, the distribution can be assumed log-normal. According to these results, geometric means can be used to characterize average values.

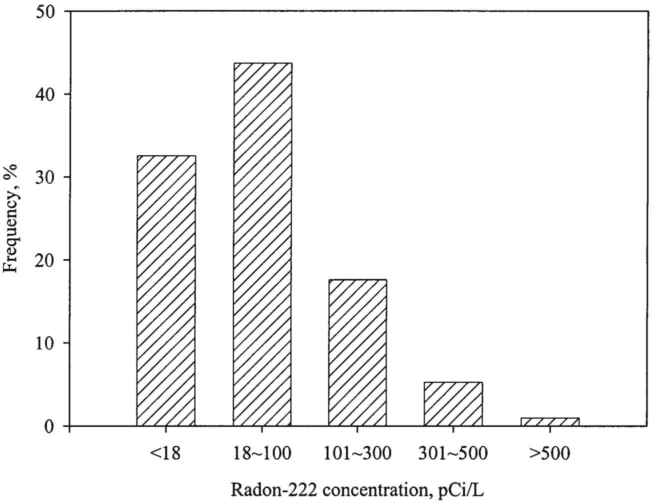
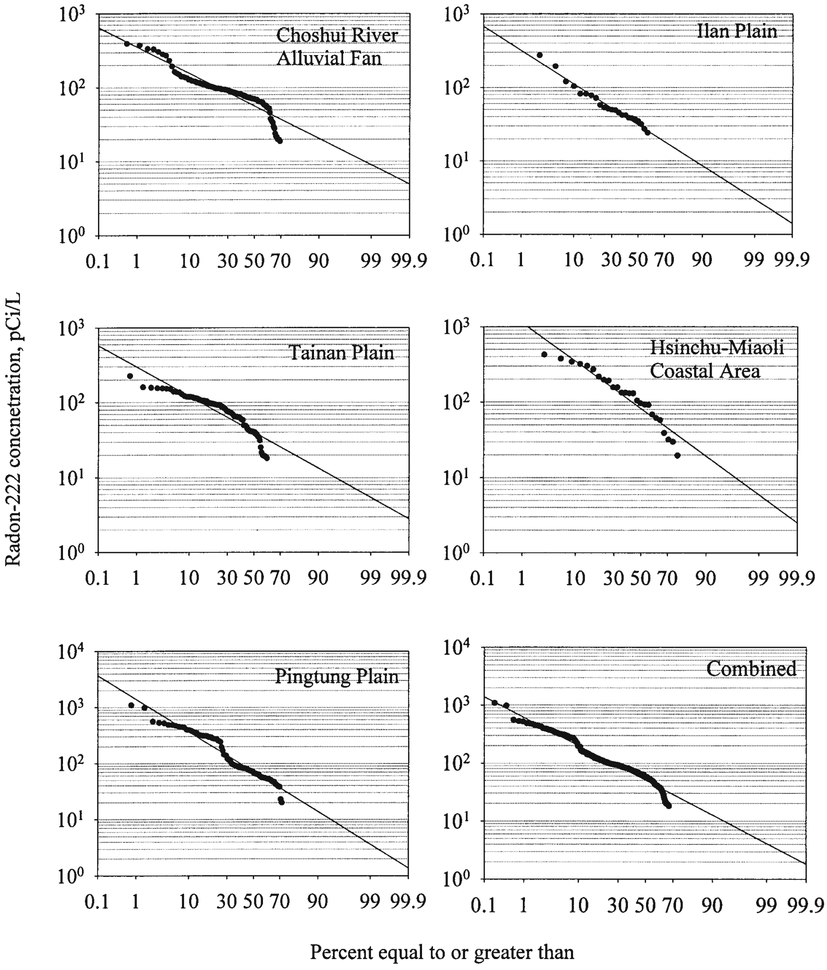


Fig. 6 Combined frequency distribution of radon-222 in five groundwater areas in Taiwan

Table 1 summarizes statistical properties of distributions of radon-222 levels for the five groundwater areas based on the 517 wells surveyed. The geometric means of radon-222 concentration are 43, 31, 61, 69, and 27 pCi/L

Fig. 7 Log-normal probability plot of radon-222 in five groundwater areas in Taiwan



DOI 10.1007/s10040-004-0384-7

Table 1 Statistical summary of radon-222 distributions for the five groundwater areas in Taiwan

Major Groundwater Area	No. of wells sampled	Radon-222 concentration, pCi/L					
		Minimum	75%-ile	Geometric Mean	Median	25%-ile	Maximum
Choshui River Alluvial Fan	175	<18	<18	43	70	96	391
Tainan Plain	142	<18	<18	31	40	91	226
Pingtung Plain	128	<18	<18	61	69	248	1,094
Ilan Plain	39	<18	<18	27	35	53	273
Hsinchu-Miaoli Coastal Area	33	<18	25	69	96	193	425
Combined	517	<18	<18	42	59	97	1,094

for Choshui River Alluvial Fan, Tainan Plain, Pingtung Plain, Hsinchu-Miaoli Coastal Area, and Ilan Plain, respectively.

Worldwide Comparison of Radon Levels in Groundwater

In the past 25 years, radon levels were measured in groundwater and spas in various countries such as Taiwan (Lin and Chen 1983; Sabol et al. 1995), Finland (Asikainen and Kahlos 1980), Norway (Banks et al. 1998a; Banks et al. 1998b), Germany (Freyer et al. 1997), Spain (Soto et al. 1995a; Soto et al. 1995b), Kenyan (Otwoma and Mustapha 1998), Saudi Arabia (Alabdula'aly 1996; Alabdula'aly 1999), and the United States (Brutsaert et al. 1981; King et al. 1982; Sasser and Watson 1978; Loomis 1987; Cech et al. 1988; Longtin 1988; Dixon and Lee 1988; Hess et al. 1985). Figure 8 summarizes worldwide radon levels in groundwater and spas reported in the literature. The geology of Taiwan is mainly composed of sedimentary rocks, therefore, the overall radon level in Taiwan groundwater is relatively low compared to other countries in the world. Because the results in Fig. 8 are for different geologic conditions in the world, data from the literature needs to be very selectively used for comparison of radon levels.

Radon-222 in groundwater has its origin in the radioactive decay of radium-226 atoms contained in rock, soil and mineral grains. Radium-226 is a member of the decay series beginning with uranium-238. Radon-222 decays by a chain of alpha and beta emitting radionuclides that leads to the stable isotope Pb-206. Asikainen and Kahlos (1979) showed that anomalously high radon-222 concentrations in groundwater in the Helsinki area were caused by high concentrations of uranium and radium-226 deposited in fissures of the bedrock. High radon-222 concentrations exist in groundwater from most granitic rocks in Maine (Brutsaert et al. 1981). Brutsaert et al. (1981) reported that granites in Maine contain 10 to 100 ppm uranium. Lin et al. (1986) reported that conglomerates and sandstones in Taiwan contain 0.87 ± 0.63 and 1.53 ± 0.59 ppm uranium, respectively. As expected, Fig. 8 reveals that the overall radon levels in Taiwan groundwater are relatively low compared to levels reported for Maine.

Loomis (1987) and King et al. (1982) reported that low radon-222 concentrations were observed in sedimentary rock aquifers in the Coastal Plain and high radon-222 concentrations were observed in granite and crystalline rock aquifers in the Piedmont in North and South Carolina, respectively. All five of the major groundwater areas of Taiwan surveyed in this study are sedimentary rock aquifers. As expected, Fig. 8 reveals that the range of radon concentrations for North Carolina and South Carolina is wider than that for Taiwan. Figure 8 also reveals that overall radon levels in Taiwan groundwater are comparable to low levels reported for North Carolina and South Carolina.

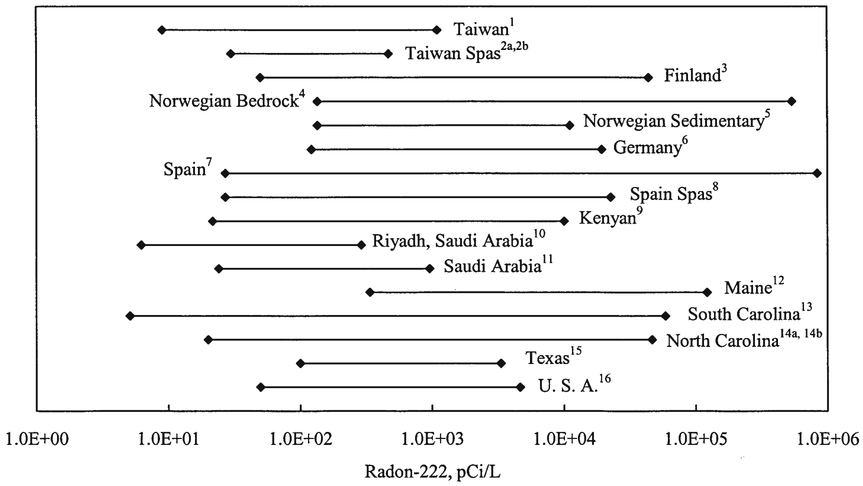


Fig. 8 Dissolved radon-222 distribution in groundwater supplies in various countries

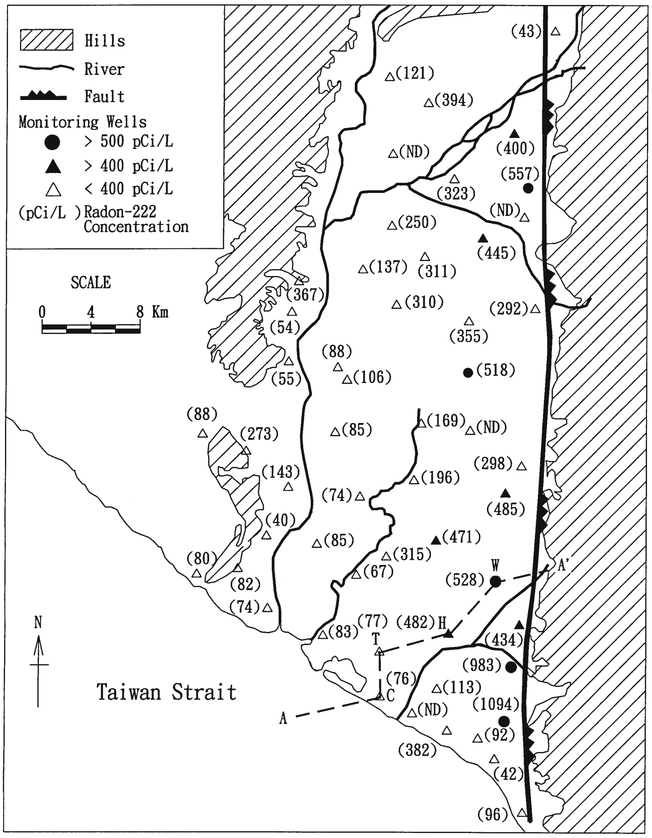


Fig. 9 Distribution of radon-222 in groundwater in the Pingtung Plain

Anomalous Concentrations of Radon in Groundwater Near Chaochou Fault

Among the 517 wells surveyed, only five wells were found with radon concentrations higher than 500 pCi/L, all located in the Pingtung Plain. The radon-222 concentration in groundwater in the Pingtung Plain (Fig. 9) varied in a wide range from below the detection limit of 18 pCi/L up to 1,100 pCi/L. However, the wells with radon concentrations higher than 400 pCi/L are all located near the Chaochou Fault. Cech et al. (1988) also reported anomalies of radon concentrations in the groundwater of Texas, USA, coinciding with geological fault systems.

Figure 10 shows radon concentrations in groundwater with the distance from the Chaochou Fault along a cross section running east-west across the Pingtung Plain. Cross section AA' correlates four well clusters, i.e., W, H, T, and C starting from the Chaochou Fault on the east side to Taiwan Strait on the west side with a total distance of about 20 km. Well cluster W is located in the area of groundwater recharge. The lithology of well cluster W is mainly gravel, which is more than 200 m thick. Recharge is mainly from rainfall and surface and subsurface channel flow. Notice also that near the Chaochou Fault in the recharge area of well clusters W and H, the radon concentration in the deep aquifer is higher than that in the shallow aquifer. For example at well cluster W, radon concentrations in the deep and shallow aquifers in the recharge area were 528 ± 43 and 276 ± 6 pCi/l, respectively. Radon concentrations in water samples taken from both surface and subsurface channel flows in the upstream catchments were also measured. Radon concentrations in surface and subsurface channel flows in the upstream

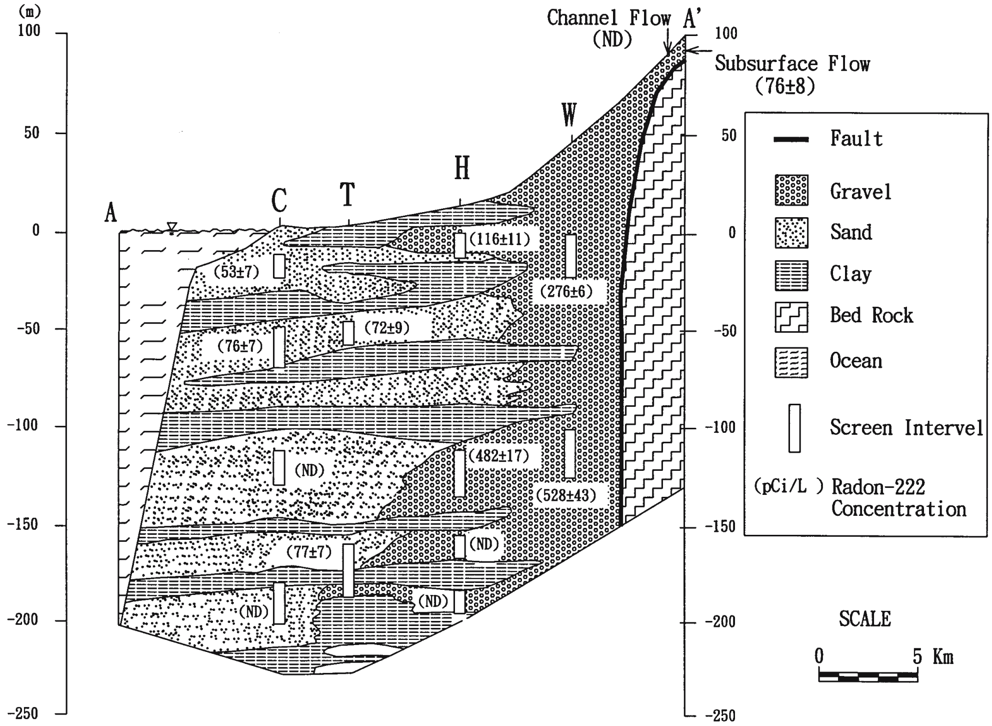


Fig. 10 Distribution of radon-222 in groundwater with the distance from the Chaochou fault along geological cross-section AA' in the Pingtung Plain

catchments were below the detection limit of 18 pCi/L and 76 ± 8 pCi/L, respectively. Based on the above observation of radon concentrations of water samples near the Chaochou Fault in the upstream catchments from both surface and subsurface channel flows, from both shallow and deep aquifers, radon appears likely coming upward from deep sources, possibly along fractures associated with the fault system. Another possibility is that the gravels located near the fault scarp may not be homogeneous from bottom to top of the sedimentary pile. Analyses of uranium-238 concentrations for the gravels are required to test the hypothesis.

Acknowledgements The support of the Water Resources Agency and Energy Commission of the Ministry of Economic Affairs, and the National Science Council of Taiwan is appreciated.

References

Alabdula'aly AI (1996) Occurrence of radon in Riyadh groundwater supplies. Health Physics 70(1):103–108

Alabdula'aly AI (1999) Occurrence of radon in the central region groundwater of Saudi Arabia. Journal of Environmental Radioactivity 44(1):85–95

Asikainen M, Kahlos H (1980) Natural radioactivity of drinking water in Finland. Health Physics 39(1):77–38

Banks D, Frengstad B, Midtgård AK, Krog JR, Strand T (1998a) The chemistry of Norwegian groundwaters: I. The distribution of radon, major and minor elements in 1604 crystalline bedrock groundwaters. The Science of the Total Environment 222(1–2):71–91

Banks D, Midtgård AK, Frengstad B, Krog JR, Strand T (1998b) The chemistry of Norwegian groundwaters: II. The chemistry of 72 groundwaters from Quaternary sedimentary aquifers. The Science of the Total Environment 222(1–2):93–105

Brutsaert WF, Norton SA, Hess CT, Williams JS (1981) Geologic and hydrologic factors controlling radon-222 in ground water in Maine. Ground Water 19(4):407–417

Cech I, Kreitler C, Prichard H, Holguin A, Lemma M (1988) Radon distribution in domestic water of Texas. Ground Water 26(5): 561–569

Dixon KL, Lee RG (1988) Occurrence of radon in well supplies. Journal American Water Works Association 80(7):65–70

Freyer K, Treutler HC, Dehnert J, Nestler W (1997) Sampling and measurement of radon-222 in water. Journal of Environmental Radioactivity 37(3):327–337

Han YL, Kuo MCT (2003) Monitoring of radon in Taiwan groundwaters. 2nd Taiwan-Japan International Workshop on Hydrological and Geochemical Research for Earthquake Prediction, Tainan, Taiwan, pp 69–71

Hess CT, Michel J, Horton TR, Prichard HM, Coniglio WA (1985) The occurrence of radioactivity in public water supplies in the United States. Health Physics 48(5):553–586

Igarashi G, Saeki S, Takahata N, Sumikawa K, Tasaka S, Sasaki Y, Takahashi M, Sano Y (1995) Ground-water radon anomaly before the Kobe earthquake in Japan. Science 269:60–61

King CY (1978) Radon emanation on San Andreas Fault. Nature (London) 271:516–519

King PT, Michel J, Moore WS (1982) Ground water geochemistry of ^{228}Ra, ^{226}Ra and ^{222}Rn. Geochimica et Cosmochimica Acta 46:1173–1182

Lin YM, Chen CC (1983) Monitoring of radon in water of Taiwan. Hoken Butsuri 18:23–27

Lin YM, Lin PH, Huang CC, Chen CJ, Chu TC (1986) Investigation of natural radionuclides in rocks of Taiwan. Nucl Sci J 23(2):138–142

Liu KK, Yeh YH, Teng TL (1986) Ground-water radon variation associated with the May 20, 1986 Hualien, Taiwan earthquake. Bulletin of the Institute of Earth Sciences, Academia Sinica (6):85–94

Longtin JP (1988) Occurrence of radon, radium, and uranium in groundwater. Journal American Water Works Association 80(7):84–93

Loomis DP (1987) Radon-222 concentration and aquifer lithology in North Carolina. Ground Water Monitoring Review 7(2):33–39

Lowry JD (1991) Measuring low radon levels in drinking water supplies. Journal American Water Works Association (4):149–153

Noguchi M (1964) Radioactivity measurement of radon by means of liquid scintillation fluid. Radioisotope 13(5):362–366

Noguchi M, Wakita H (1977) A method for continuous measurement of radon in groundwater for earthquake prediction. Journal of Geophysical Research 82:1353–1357

Otwoma D, Mustapha AO (1998) Measurement of ^{222}Rn concentration in Kenyan groundwater. Health Physics 74(1):91–95

Prichard HM, Gesell TF (1977). Rapid measurements of ^{222}Rn concentrations in water with a commercial liquid scintillation counter. Health Physics 33:577–581

Prichard HM, Venso EA, Dodson CL (1992) Liquid-Scintillation analysis of ^{222}Rn in water by alpha-beta discrimination. Journal of Radioactivity and Radiochemistry 3(1):28–36

Sabol J, Weng PS, Mao CH (1995). Monitoring of ^{222}Rn in Taiwanese hot spring spa waters using a modified electret ion chamber method. Health Physics 68(1):100–104

Sasser MK, Watson JE (1978) An evaluation of the radon concentration in North Carolina ground water supplies. Health Physics 34:667–671

Soto J, Fernández PL, Quindós LS, Gómez-Arozamena J (1995a) Radioactivity in Spanish spas. The Science of the Total Environment 162:187–192

Soto J, Fernández PL, Gómez J, Ródenas C (1995b) Study of the occurrence of ^{222}Rn and ^{226}Ra in drinking water in Spain. Health Physics 69(6):961–965

United Nations Scientific Committee on the Effects of Atomic Radiation (1988) Sources, Effects and Risks of Ionizing Radiation. Report to the General Assembly, United Nations, New York, pp 24–79