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Continuous monitoring of groundwater radon for evaluating chemical and structural properties and fluid flow variations of shallow aquifer systems

By

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with 5 Figures

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Abstract: We have carried out continuous monitoring of radon concentration in groundwater at eight wells in Nishinomiya City, Hyogo Prefecture, Japan. The shallowest major aquifer, which we call the first aquifer, extends widely at a depth about 3-5 meters in the region. We pay our major attention to the first aquifer, because it provides us with abundant high quality water, which has long been utilized for brewing Sake. We made the radon monitoring at 5 wells tapping the first aquifer. The first aquifer can be characterized by very high radon concentration and its large temporal variation. Time-averaged values of radon concentration at 4 shallow wells were about 70-80 Bq/l, which is almost the highest value ever reported for natural water throughout the Japanese Islands. These high radon concentrations can be attributed to uranium-rich sediments in the aquifer, which had been brought from the Rokko Mountains region on the north. Large temporal variations of radon concentration observed at the shallow wells probably reflect the heterogeneous distribution of the grain size of sediments, which makes the groundwater flow unstable. We examined radon concentrations in deeper aquifers at three wells with depths of 8-17 m for comparison. Absolute values of radon concentrations and their temporal variations at the deeper wells are smaller than those in the first aquifer, which suggests the small contributions of uranium-rich sands to the sediments compared with the first aquifer. The radon concentration at a well with a depth of 16 m showed periodical variations responding to the ocean tide, which demonstrates that very small signals of crustal deformations can be detected by the continuous monitoring of groundwater radon.

Contents

I. Introduction

Radon (^{222}Rn) has served as a useful tracer in various fields of earth sciences, such as earthquake prediction (cf., King 1986) and ocean circulation (cf., Broecker and Peng, 1982). ^{222}Rn is a radioactive nuclide, with a half life about 3.8 days, belonging to the decay series of ^{238}U (Fig. 1). Because radon is an inert gas which is highly soluble in water, its behavior is simple and can be understood on the basis of physical processes such as dissolution, adsorption, diffusion and fluid advection, as well as the radioactive decay (cf., Ozima and Podosek, 1983). Therefore, radon has been utilized as a tracer for flow, circulation, and mixing of natural water in various circumstances.

Since the continuous monitoring system of groundwater radon was developed by Noguchi and Wakita (1977), extensive radon monitoring has been operated mainly by University of Tokyo and Geological Survey of Japan for the purpose of earthquake prediction for about 20 years. A number of anomalous changes associated with earthquakes have been reported (e.g., Wakita et al. 1980; Igarashi and Wakita 1990), which strongly support that groundwater radon is a sensitive tracer for the crustal strain changes associated with earthquake occurrences. However, the mechanisms causing such earthquake-related radon changes are still poorly understood.

The radon concentration in groundwater is basically proportional to the uranium concentration in adjacent rocks in an aquifer. However, because of the very short recoil length of radon (about 3×10^{-8} m), only a small portion of radon atoms produced in the rocks will be released to the surrounding groundwater; except for those produced at the near-surface area of rock grains, radon atoms cannot escape from rock grains. Therefore, the radon concentration in groundwater is largely dependent on, and roughly negatively proportional to, the effective grain size of rocks in an aquifer (Torgersen et al., 1990). For example, micro-crack formations due to compressional stress will reduce the effective grain size of rocks, which may result in enhancements of radon concentration in the groundwater. Thus the groundwater radon concentration is expected to reflect not only chemical but also structural properties of rocks in the aquifer.

Radon atoms released from rocks to the surrounding groundwater will be transported along with the flow of groundwater. The flow of groundwater is generally very slow, from several centimeters to several meters a day. Therefore, radon atoms can move a very limited length during their life times, less than several tens of meters. Accordingly, temporal variations of groundwater radon concentration are probably caused by local changes in chemical, structural, or fluid flow properties restricted in a very small area near an observation well, even if such local changes would be induced by changes in tectonic stress field extending to a much larger area.

To investigate such local phenomena which may directly result in groundwater radon changes, it is necessary to monitor groundwater radon intensively at several wells within a small area. In the present paper, we report preliminary results of an intensive monitoring of groundwater radon in Nishinomiya City, Hyogo Prefecture, Japan.

Acknowledgments

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II. Radon observation system

We have measured groundwater radon using a system developed by Tasaka and Sasaki (1992). Groundwater is continuously introduced into a detection chamber with a flow rate of about 1 l/min. Radon dissolved in groundwater is degassed to the gas phase in the chamber. A container of electrostatic collector is mounted just above the interface between gas and liquid phases in the chamber. The container is equipped with an alpha ray detector of the PIN photodiode (Hamamatsu Photonics K.K.) with a surface area of 1 cm^2. Since a static voltage of -120 V is applied between the PIN photodiode and the bottom of the container, positive ions of daughters of ^{222}Rn such as ^{214}Po and ^{218}Po (Fig. 1) are collected on the surface of the PIN photodiode. Then alpha rays emitted mainly by the decays of ^{214}Po and ^{218}Po are detected as electric currents through the PIN photodiode. The electric currents are amplified and then counted with a high speed 256 channels AD converter, which is controlled by a Z80 microcomputer system. The data of the alpha ray counting are transferred to a personal computer through an RS232C port. A detailed description of the system is given by Tasaka and Sasaki (1992).

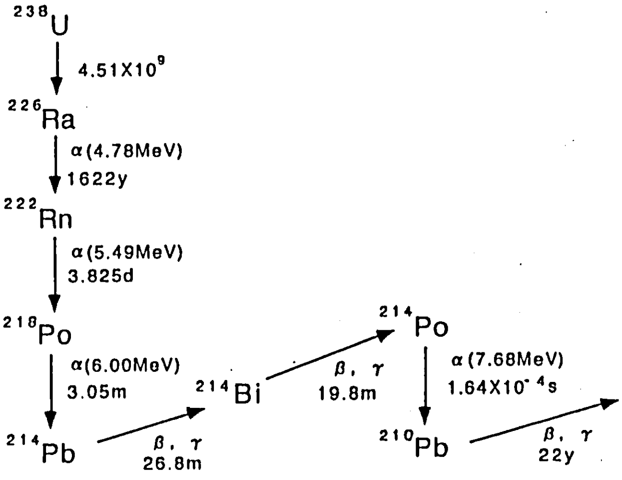

Fig.1. Radioactive decay scheme of ^{222}Rn, alpha ray energies, and half lives.

III. Observations at Nishinomiya City

We have carried out continuous monitoring of groundwater radon at eight observation wells, which are scattered in Nishinomiya City, Hyogo Prefecture (Fig. 2). This area is located on the south of the Rokko Mountains and is about 1-2 km distant from Seto Inland Sea. A major aquifer, situated at a depth of about 3-5 meters, is filled mainly with granitic sands and rocks which are considered to have been transported from the Rokko Mountains area. This aquifer, which we call the first aquifer, has very high permeability, above $5x10^{-2}$ cm/s throughout the layer (Sumikawa, 1990), providing us with abundant groundwater resources. Furthermore, the groundwater from the first aquifer is rich in phosphorous, potassium, and calcium (Sumikawa, 1990), which makes the water suitable for brewing Sake, the Japanese traditional liquor. Traditional Sake breweries have long been using this groundwater. The location of a major group of breweries in this area is shown in Fig.2.

To preserve the quality of this groundwater, over 60 wells have been bored within a small area less than 10 km^2 surrounding the breweries and periodic examinations of chemical compositions of groundwater have been continued for about 40 years (Sumikawa, 1990). Thus the environmental conditions in this area is suitable for investigating the local variations of groundwater radon. As the first step of our researches, we made continuous monitoring of radon concentration in the groundwater from the first aquifer at 5 different wells for the periods from 4 days to 12 days. Unfortunately, we could prepare only one instrument for radon monitoring this time. So we were obliged to make a single radon measurement repeatedly, by moving the instrument from one well to another.

In addition, we also made radon monitoring at three wells tapping deeper aquifers for comparison: two wells with depths of 16 m and 17 m tapping the second major aquifer, which has very high permeability comparable to that of the first aquifer (Sumikawa, 1990), and one well with a depth of 8 m tapping a less permeable minor aquifer between the first and second major aquifers.

IV. Results and discussion

Fig. 3 and Fig. 4 show the results of continuous monitoring of groundwater radon in Nishinomiya City. Not only the absolute values of radon concentration, their temporal variation patterns are also surprisingly different from well to well. Such differences are considered to reflect variable flows of the groundwater as well as inhomogeneous distributions of uranium concentration and grain size of sediments in the aquifers. Thus the radon concentration will provide valuable information about the characteristics of the aquifer systems. However, to exactly characterize both the spacial and temporal variations of radon concentration in groundwater in this area, our observations are insufficient and further observations at more wells for longer periods should be needed. Nevertheless, several interesting features have already been revealed as follows.

A. The first aquifer

Absolute values of radon concentration at the shallow wells are very similar to one another, except for that at HM-1; time-averaged values of radon concentrations at S-1, H-5, H-6, and 3H-5 are 70-80 Bq/l, and that at HM-1 is 15 Bq/l (Bq is a unit for the number of radioactive decays per unit time, 1 Bq corresponds to 1 decay per second). Ishii and Horiuchi (1992) made a wide survey of the radon concentration in natural water all over the Japanese Islands; 35 natural waters sampled from various regions of Japan range from 0.24-98.91 Bq/l, with a mean value of 12.98 Bq/l. Thus the radon concentrations at the shallow wells, except for HM-1, are

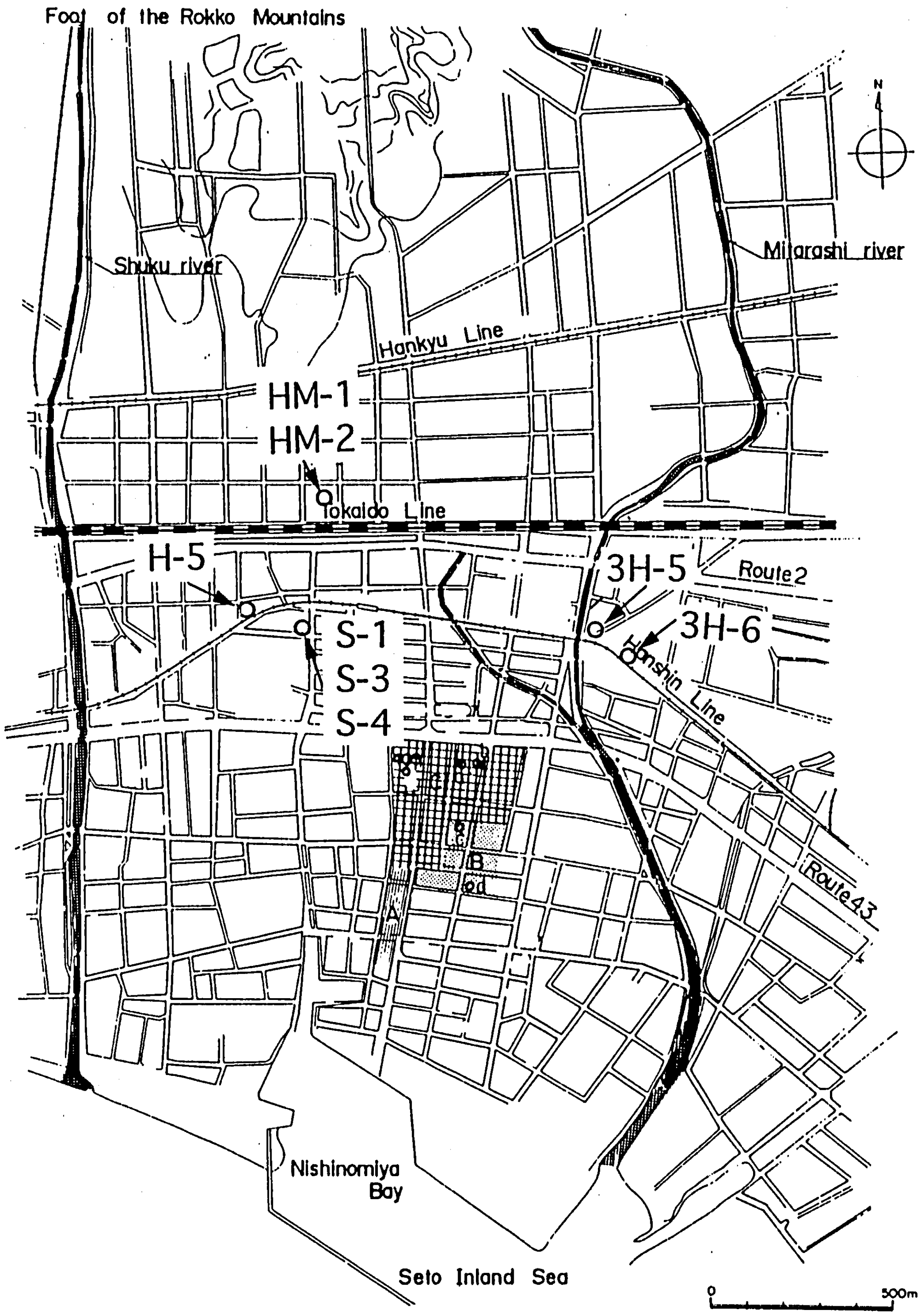

Fig. 2. Location map of the observation wells and the Sake breweries in Nishinomiya City. Sake breweries until about 1920 (A), 1920-1933 (B), and the present (C). Modified from Fig.4. in Sumikawa (1990).

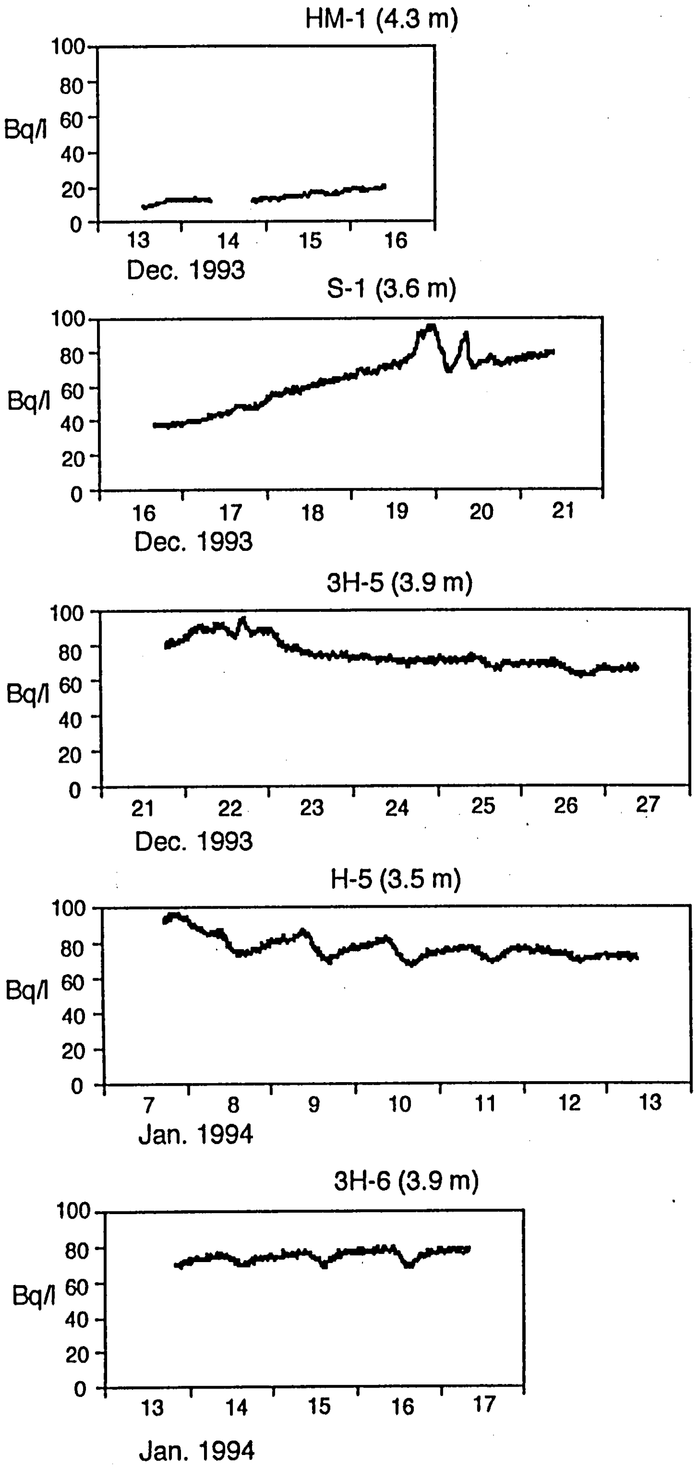

Fig.3. Radon concentrations at the shallow wells tapping the first aquifer. Depths of the wells are shown in parentheses.

almost the highest value of Japan, whereas that at HM-1 is much smaller and close to the mean value of natural water in Japan.

According to Sumikawa (1990), sediments from the aquifer layers at the S-1, H-5, H-6, and 3H-5 wells are dominated by light coloured granitic sands, whereas those from the aquifer at HM-1 are mainly composed of dark coloured marine sediments. Sumikawa (1990) examined the distribution of water-head gradients of groundwater and found that there are several major groundwater flow lines from north to south in this area. He further found out that the present groundwater flows are closely related to the positions of ancient rivers. Thus sediments in the first aquifer were mainly transported along with these ancient river flows from the Rokko Mountains area. The S-1, H-5, H-6, and 3H-5 wells are situated at the major groundwater flows from north to south, where a large amount of granitic sands rich in uranium were brought. Hence, the high radon concentrations at these wells can be attributed to large contributions of uranium-rich granitic sands to the sediments. On the other hand, the HM-1 well is distant from the major groundwater flows and its aquifer is filled with marine sediments relatively poor in uranium. This is the reason why the radon concentration at HM-1 is much less than the other shallow wells. Thus the difference in radon concentration of groundwater is consistent with the chemical composition and origin of sediments in the aquifers.

It should be noted that the radon concentrations at the shallow wells show large temporal variations. Not only uranium concentration but the grain size of sediments in an aquifer also have large influence on radon concentration of groundwater (Torgersen et al., 1990). As mentioned above, uranium-rich granitic sands which were transported along with the flows of ancient rivers play an important role in enhancing radon concentrations in the first aquifer. It is likely that heavy rainfalls would have caused floods of the ancient rivers repeatedly. Such flood events would have brought large grained rocks intermittently to this area, which caused heterogeneity in the grain size of sands and/or rocks in the first aquifer. Hence, the observed large temporal variations of groundwater radon concentration in the first aquifer probably reflect the heterogeneous distribution of the grain size of sediments, which can be explained by the formation process of the aquifer.

Furthermore, the irregular variations of radon concentration, which are particularly notable in the wells S-1 and 3H-5, strongly suggest that fluid flow in the first aquifer is considerably unstable. The unstable fluid flow is also attributable to the heterogeneous distribution of the grain size of sediments. In general, the groundwater flow is determined by the distribution of water-head gradient

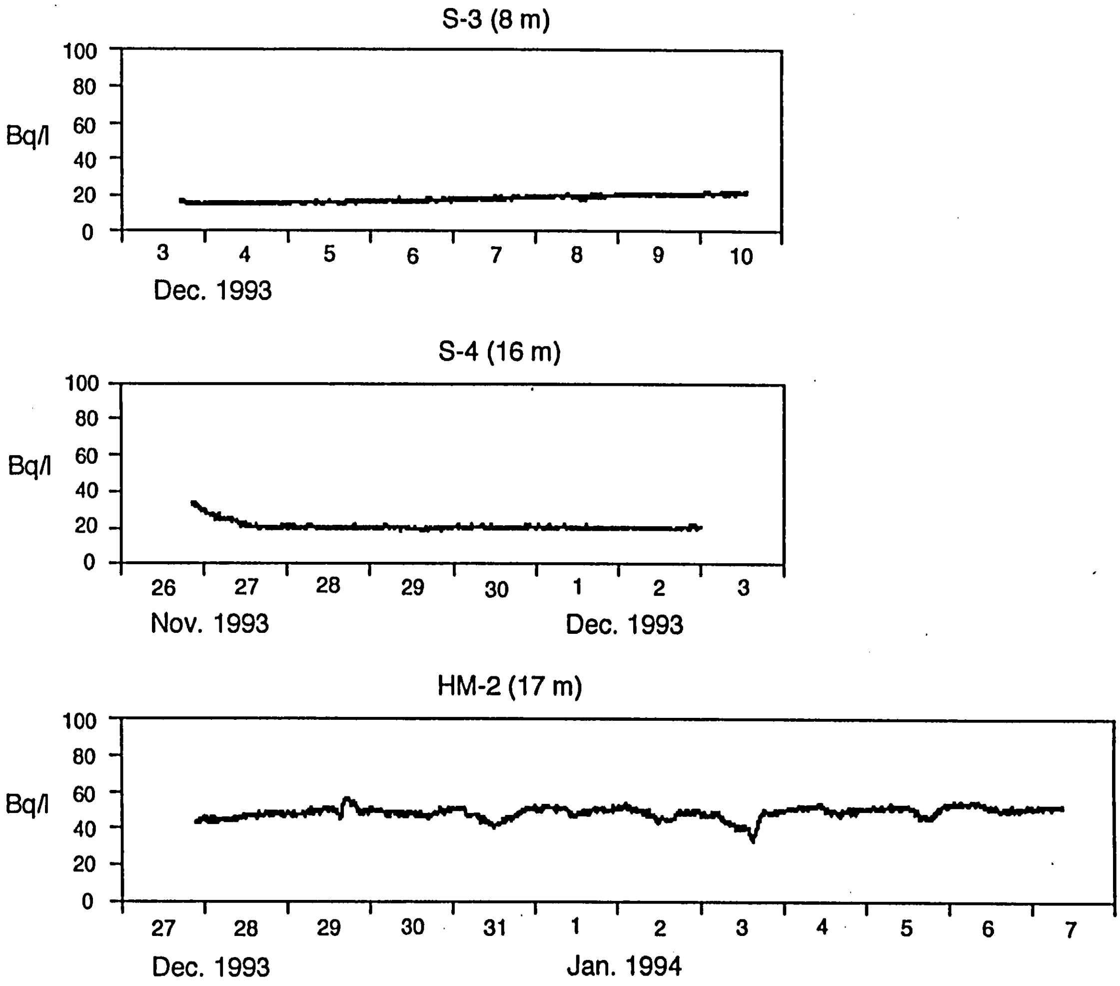

Fig.4. Radon concentrations at deeper wells. Depths of the wells are shown in parentheses.

and permeability of an aquifer. The grain size of sediments is the most important parameter controlling the permeability of an aquifer.

At the H-5 and 3H-6 wells, daily variations of radon concentration are dominant, which suggest that human activities affect the fluid flow significantly. This implies that because of its heterogeneous structure, very small changes in fluid flow can easily be detected as large variations of radon concentration. Thus continuous monitoring of groundwater radon is a sensitive tracer for fluid flow variations particularly of shallow heterogeneous aquifers.

B. Deeper aquifers

Absolute values of radon concentration at the deeper wells are small compared with those at the shallower wells; time-averaged values are 20 Bq/l at S-3 and S-4, and 50 Bq/l at HM-2. In addition, radon concentrations at the deeper wells are much more stable than those in the shallower wells. These differences will be closely related to chemical and structural differences of the aquifer systems.

The S-3 well taps a minor aquifer, with permeability

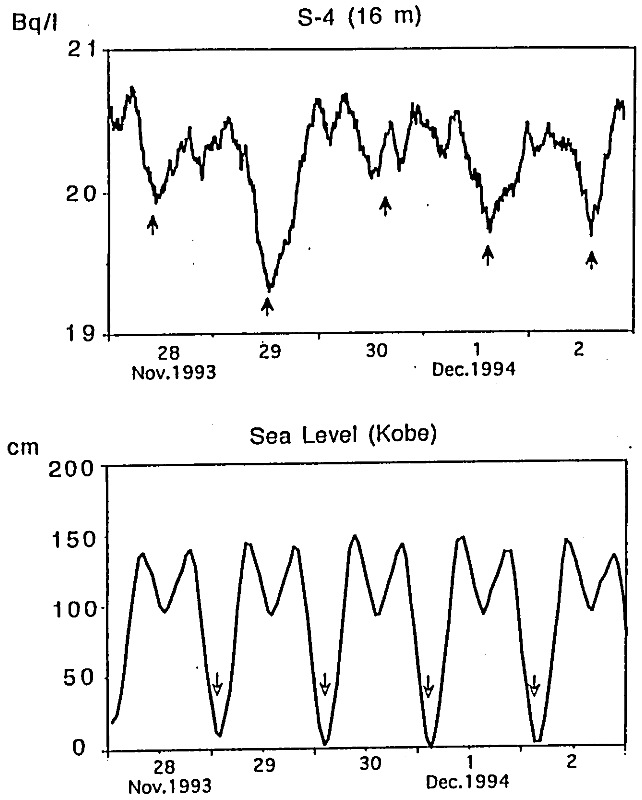

Fig. 5. An expanded plot of the radon concentration at the S-4 well and the sea level at the Kobe harbor. The data on radon concentration are averaged for time intervals of 3 hours to eliminate the noises with shorter periods.

less than 5×10^{-2} cm/s, situated between the first and second major aquifer. This layer is mainly composed of fine grained marine sediments whose uranium concentration is probably much less than the sediments at the first aquifer. As a result, the radon concentration at the S-3 well is only about a quarter of the S-1 well, although the waters at these two wells are taken from the places only about 4 m vertically, and 2 m horizontally distant from each other. Thus there exists a striking vertical contrast of groundwater radon concentration in this area.

Two wells tapping the second major aquifer, S-4 and HM-2, which are about 400 m distant from each other, show quite different features of radon concentration both in their absolute values and temporal variations. Such differences are found at the wells tapping the first aquifer, between S-1 and HM-1. This may imply that the chemical and structural properties of the second major aquifer also vary considerably in the north to south direction. Further observations will be needed to clarify the horizontal distribution of radon concentration in the second aquifer.

The radon concentration at the S-4 well is extremely stable, which suggests that the sediments in the aquifer is chemically and structurally homogeneous and that the groundwater flow is also stable. However, a closer examination of the data has revealed interesting periodical variations. Fig. 5 plots the radon concentration at S-4 which is averaged for time intervals of 3 hours to eliminate noises with shorter periods. The data on the sea level at Kobe harbor are shown in Fig. 5 for comparison. The variations of the radon concentration at S-4 is quite similar to that of the sea level; variations with a period about 25 hours are dominant and smaller variations with a period about 12.5 hours can also be noted. The variation of radon concentration lags about 12 hours behind that of the sea level. Thus it has turned out that the radon concentration at the S-4 well responds clearly to the ocean tide. Changes in the loading of sea water due to the ocean tide will cause the periodical deformation of the crust. Such deformations due to the ocean tide are the order of 10^{-8} in volumetric strain (Sato and Hanada, 1984). Hence, our observation demonstrates that by the continuous monitoring of groundwater radon, we can detect signals of very small crustal deformations.

V. Conclusions

We report preliminary results of continuous monitoring of groundwater radon in Nishinomiya City, Hyogo Prefecture, to investigate detailed chemical and structural properties and fluid flow variations of shallow aquifer systems. Both of the absolute values and their temporal variations show large differences from well to well, reflecting the characteristics of the aquifer systems.

(1) Absolute values of radon concentration at the shallow wells are extremely high and are almost the highest value of natural water in Japan, which can be attributed to large contributions of uranium-rich granitic sands to the sediments brought from the Rokko Mountains region on the north.

(2) The radon concentrations at the first aquifer show large temporal variations, which probably reflect the heterogeneous distribution of the grain size of sediments and unstable flows of groundwater. It is likely that the heterogeneous distribution of the grain size of sediments were caused by the floods of ancient rivers which had brought granitic rocks of various sizes intermittently to this area.

(3) Some of the shallow wells show daily variations of radon concentration, which suggests that human activities affect the fluid flow significantly. Continuous monitoring of groundwater radon is a sensitive tracer for fluid flow variations particularly of such shallow heterogeneous aquifers.

(4) Absolute values of radon concentration at the deeper wells are much less than those in the first aquifer, which suggests a striking vertical contrast of groundwater radon concentration in this area. Furthermore, two wells tapping the second major aquifer, which are about 400 m distant from each other in the north to south direction, show quite different features of radon concentration both in their absolute values and temporal variations. This may imply that the chemical and structural properties of the second major aquifer vary considerably in the north to south direction.
(5) Periodical variations of radon concentration responding to the ocean tide was detected at a well with a depth of 16 m, which demonstrates that by the continuous monitoring of groundwater radon, we can detect signals of very small crustal deformations.

We are now operating 4 instruments for radon monitoring in the Nishinomiya City. The new simultaneous monitoring of groundwater radon at multi-stations will provide much more information about the complicated aquifer systems.

References

Broecker, W.S. and Peng, T.-H., 1982: *Tracers in the sea*. pp. 690. The Lamont -Doherty Geological Observatory.

Igarashi, G. and Wakita, H., 1990: Groundwater radon anomalies associated with earthquakes. *Tectonophys.*, **180**, 237-254.

Ishii, T. and Horiuchi, K., 1992: Radon in natural water. *Environmental radon*, 160-166.

King, C.-Y., 1986: Gas geochemistry applied to earthquake prediction: an overview. *J. Geophys. Res.*, **91**, 12269-12281.

Noguchi, M. and Wakita, H. 1977: A method for continuous measurement of radon in groundwater for earthquake prediction. *J. Geophys. Res.*, **82**, 1353-1357.

Ozima, M. and Podosek, F.A., 1983: *Noble Gas Geochemistry*. pp. 367. Cambridge University Press.

Sato, T. and Hanada, H., 1984: A program for the computation of oceanic tidal loading effects 'GOTIC'. *Publ. Int. Latitude Obs. Mizusawa*, **18**, 29-47.

Sumikawa, K., 1990: On the groundwater of Nishinomiya district, Kinki, Japan -with special reference to the characteristics of permeability of the aquifer and chemical composition of the Miyamizu-. *J. Sci. Hiroshima Univ.*, **9**, 361-376.

Tasaka, S. and Sasaki, Y., 1992: Observations of underground water radon concentration at the Kamioka Mine, Gifu Prefecture. *Zisin*, **45**, 1-9.

Torgersen, T., Benoit, J. and Mackie, D., 1990: Controls on groundwater Rn-222 concentrations in fractured rock. *Goephys. Res. Lett.*, **17**, 845-848.

Wakita, H., Nakamura, Y., Notsu, K., Noguchi, M. and Asada, T., 1980: Radon anomaly: A possible precursor of the 1978 Izu-Oshima-Kinkai earthquake. *Science*, **207**, 882-883.

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