DETECTION OF THE FLOWING SECTIONS OF GROUNDWATER MEASURING DISSOLVED RADON BY AN AQUATIC ALPHA TRACK METHOD

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ABSTRACT

Dissolved radon is a good tracer to track the movement of groundwater in a stratum. Radon logging is a very useful technique for detecting groundwater flow in a boring well, but radon requires prompt measurement, because all isotopes of radon have short half-lives. However, prompt measurement of radon concentration is very difficult in the field, because it takes several hours or days for the extraction from groundwater and counting radioactivity in a laboratory after sampling. In this study, a method for in-situ measurement of radon was developed without sampling groundwater. The in-situ method uses the alpha track method, which is commonly used to measure radon concentrations in air, to measure radon dissolved in groundwater. Since alpha particles have a very short range of a few tens of micrometers in water, the alpha track method is not usually used to measure dissolved radon concentration. However, to date, no practical solid detector has been produced to measure radon dissolved into water. If an alpha track detector that measures radon dissolved in water can be produced, the short range of alpha particles emitted from radon and its daughters in water will conversely greatly help to trace groundwater movement because the dissolved radon and its daughters can not make tracks on the surface of a solid detector without radon directly touching the surface. Furthermore, this new aquatic alpha track method does not require prompt measurement, because radon concentration is left on the surface of detectors as the integrated numbers of tracks in proportion to the length of time the solid detector has been exposed in the water. Consequently, this method will be useful for tracking the in-situ groundwater movement in groundwater surveys or earthquake prediction.

INTRODUCTION

Groundwater movement is one of the key factors in site selection for the disposal of high level radioactive waste. Tracers are useful for detecting groundwater movement in a geological stratum, and various tracers have been used to survey groundwater flow. Some tracers such as salt or dye were artificially introduced into aquifers to investigate the location of groundwater discharge or the routes of groundwater. Recently, some (stable or radioactive) isotopes in nature have been used in regional and local groundwater surveys as a natural tracer. The typical survey method is dating of groundwater by using natural radionuclides of ^{3}H, ^{14}C, ^{36}Cl and so on. These radionuclides have relatively long half lives. On the other hand, there are also many natural radionuclides which have short half lives, such as radon.

Since radon is a radioactive noble gas, it is an ideal tracer to track groundwater. However, the longest half life, 3.8 days for ^{222}Rn is usually too short for convenient measurement . The short half life is a major problem for groundwater surveys, because groundwater moves generally very slowly. On the other hand, it is a useful advantage for detecting the circulation of groundwater in a small local area so that all isotopes of radon have a short half life. For example, Hoehn and Guten (1989) succeeded in evaluating the effects of intrusion of surface water into an aquifer. Furthermore, radon anomalies in groundwater are considered to be a good indicator of imminent earthquakes (King, 1986, Igarashi et al., 1995). These anomalies have not accumulated long-term changes because ^{222}Rn decays away in about three weeks.

A short half life is a disadvantage in field observations for measuring dissolved radon concentration after sampling groundwater. A conventional technique for determining radon concentration in water involves intricate extraction of it into a solvent solution before counting with a scintillation counter. In the field, it is impossible to perform this sensitive chemical operation promptly and to count radioactivity by using a precise analytical machine. Consequently, in this study, we developed an in-situ technique counting radon dissolved into water and demonstrated that it possible to distinguish between permeable and impermeable fissures by using this new technique in field observations.

MATERIALS AND METHODS

We prepared a polycarbonate plate of 1 mm thickness with a vaporized carbon film as a solid detector for α tracks in water. The thickness of the carbon film ranges from 15 to 30 *m*m and efficiently leaves tracks produced with α particles emitted from the dissolved radon (^{222}Rn) and its daughters (^{218}Po and ^{214}Po) on the surface of the detector. The carbon film adsorbs the roving ^{222}Rn, ^{218}Po and ^{214}Po in water on the surface of the detector and absorbs the energy of α particles, leaving tracks on the surface of the polycarbonate.

Alpha tracks are counted by using a microscope after chemical etching. The polycarbonate plate was etched by soaking in a solution of 6N NaOH at 70°C for 90 mins. The carbon film protects from etching other areas on the plate without tracks damaged with α particles emitted from radon (^{222}Rn) and its daughters (^{218}Po and ^{214}Po) during soaking in a strong alkali solution.

We investigated the correlation between the density of α tracks left on the solid detector (polycarbonate plate) and concentration of radon (^{222}Rn) and its daughters (^{218}Po and ^{214}Po) dissolved in water by the following procedures. We soaked solid detectors with the thin carbon film in the radon solution secular equilibrated with its daughters. We measured the inverse relation between decrease of the α track density and the elapsed of time of the test. We exchanged solid detectors in the radon solution once a day following the predetermined schedule i.e. 1, 2, 3, 4, 5, 7 and 8 days after starting the exposure test. We confirmed the correlation between the density of α tracks and the concentration of dissolved radon and its daughters emitting α-particles.

We used this method for a field investigation to survey a section in which groundwater was moving in a boring well. The scheme for determining the section in the boring well with moving groundwater is shown in Fig. 1. We measured the density of α tracks for one week exposing polycarbonate detectors with thin carbon film to groundwater in boring wells. The detectors (20x20 mm) inserted into Teflon cases were attached to a stainless steel wire cable at 1-meter intervals. The recovered detectors were etched into a hot-6N-NaOH solution for 90 mins., after recovering detectors from boring wells, and washing the surface of the detector gently but thoroughly with distilled water.

STUDY AREA

The groundwater survey was conducted at the site shown in Fig. 2. The geological stratum and distribution of faults and fissures are shown in Fig. 3. This site was formed with Cretaceous granite and granite gneiss rocks as bedrock. These rocks are characterized with many fissures. Groundwater flow has been controlled by three different groups of fissures, which are categorized as follows.

Single fissure (SF)

This is an independent fracture and is generally closed with filling minerals, which are calcite, zeolite and clay minerals. The filled fracture is impermeable, but the weathered one is highly permeable.

Dense fracture-developing zone with parallel orientation (PF)

The fracture zone is formed with a high density of parallel fractures extending in the same direction. It is generally a highly permeable zone. The filling minerals are deposited on the surface of fractures.

Cataclasite zone (CTZ)

This is a fracture zone in which fractures are fully consolidated by filling clay minerals or breccia. Therefore, it is usually impermeable. However, since the surroundings of CTZ develop tiny fissures, it is potentially permeable.

Consequently, shallow groundwater which is recharged by precipitation flows from the high mountainous regions to the sea following a topographical slope through the aforementioned different fissure systems, due to the tidal effects.

RESULTS AND DISCUSSIONS

(1) Theoretical consideration of density of α-tracks

We theoretically estimate the total alpha particles emitted from radon and its daughters dissolved into water assuming that: a) initially radon only dissolves into water without any daughters, b) daughters are produced following alpha-decay of radon in water, c) radon and its daughters decrease by radioactive decay and are not lost by volatizing, adsorbing and precipitating in water. Furthermore, we consider the only the decay chain from ^{222}Rn to ^{214}Po.

Initially, the number of atoms of ^{222}Rn dissolved in water is N_0. Atoms of ^{222}Rn decrease following the decay formula and atoms of ^{218}Po and ^{214}Po increase as follows (Bateman,1910):

$$^{222}Rn \rightarrow \frac{dN_1}{dt} = -l_1 N_1 \rightarrow N_1 = N_0 e^{-l_1 t} \qquad (1)$$

$$^{218}Po \rightarrow \frac{dN_2}{dt} = l_1 N_1 - l_2 N_2 \rightarrow N_2 = N_0 \frac{l_1}{l_2 - l_1}(e^{-l_1 t} - e^{-l_2 t}) \qquad (2)$$

$$^{214}Po \rightarrow \frac{dN_2}{dt} = l_3 N_3 - l_4 N_4 \rightarrow N_4 = N_0 l_1 l_2 l_3 \{\frac{e^{-l_1 t}}{(l_2 - l_1)(l_3 - l_1)(l_4 - l_1)}$$
$$+ \frac{e^{-l_2 t}}{(l_1 - l_2)(l_3 - l_2)(l_4 - l_2)} + \frac{e^{-l_3 t}}{(l_1 - l_3)(l_2 - l_3)(l_4 - l_3)} + \frac{e^{-l_4 t}}{(l_1 - l_4)(l_2 - l_4)(l_3 - l_4)}\} \qquad (3)$$

The total alpha particles emitted in water during (t_1 - t_2) from time t_1 to time t_2 is given by integrating eq. (1), eq. (2) and eq. (3) from time t_1 to time t_2 :

$$^{222}Rn \rightarrow \int_{t_1}^{t_2} l_1 N_1 dt = N_0 (e^{-l_1 t_1} - e^{-l_1 t_2}) \qquad (4)$$

$$^{218}Po \rightarrow \int_{t_1}^{t_2} l_2 N_2 dt = N_0 \{ \frac{l_2}{l_2 - l_1} (e^{-l_1 t_1} - e^{-l_1 t_2}) - \frac{l_2}{l_2 - l_1} (e^{-l_2 t_1} - e^{-l_2 t_2}) \qquad (5)$$

$$^{214}Po \rightarrow \int_{t_1}^{t_2} l_4 N_4 dt = N_0 l_1 l_2 l_3 l_4 \{ \frac{(e^{-l_1 t_1} - e^{-l_1 t_2})}{l_1 (l_2 - l_1)(l_3 - l_1)(l_4 - l_1)} + \frac{(e^{-l_2 t_1} - e^{-l_2 t_2})}{l_2 (l_1 - l_2)(l_3 - l_2)(l_4 - l_2)}$$
$$+ \frac{(e^{-l_3 t_1} - e^{-l_3 t_2})}{l_3 (l_2 - l_3)(l_1 - l_3)(l_4 - l_3)} + \frac{(e^{-l_4 t_1} - e^{-l_4 t_2})}{l_4 (l_2 - l_4)(l_3 - l_4)(l1 - l_4)} \} \qquad (6)$$

Where, λh_1, λ_2, λ_3, λ_4 are decay constants of ^{222}Rn, ^{218}Po, ^{214}Bi and ^{214}Po, respectively.

Since λ_1, λ_2, λ_3, λ_4 are 2.0×10^{-6}/sec, 3.8×10^{-3}/sec, 8.5×10-4/sec, 4226/sec, respectively, and λ $\lambda_2/(\lambda_2-\lambda_1)$ and $\lambda_2\lambda_3\lambda_4/(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)$ in eqs. (5) and (6) are approximately 1, and since other terms are negligible compared to 1, the number (M) of total alpha particles emitted into water during (t_1 - t_2) are given by:

$$M = 3N_0 (e^{-l_1 t_1} - e^{-l_1 t_2}) \qquad (7)$$

The density of alpha tracks left on a solid detector is proportional to the total alpha particles emitted from dissolved radon and its daughters during (t_1 - t_2). If ^{218}Po and ^{214}Po are initially equilibrated with ^{222}Rn in water, the contribution of alpha particles emitted from the initial sources of these daughters must be corrected when counting total alpha particles. If these daughters are more readily adsorbed on the solid detector with carbon film compared to radon, we must correct the total density of α-tracks given from them even if their initial sources are negligible.

(2) Measurement of density of α-tracks in water by using the aquatic α-track method

Figure 4 shows the correlation between decrease in density of α--tracks and increase in elapse of time in the exposure test in distilled water equilibrated with ^{222}Rn. Since the ^{222}Rn solution was prepared by soaking a ^{226}Ra source in distilled water for three weeks, excess ^{218}Po and ^{214}Po exist at the start of the test as initial sources in the solution that are not considered in the aforementioned theoretical consideration. For the first three days, the density of α-tracks decreased following a decay curve with an apparent half life of 0.8 days, probably due to ^{222}Rn 's daughters ^{218}Po and ^{214}Po compared with 3.8 days of ^{222}Rn, because ^{218}Po and ^{214}Po as initial sources, which are α-emitters and are released constantly ^{222}Rn from ^{226}Ra, not only have very short half-lives of 5.05 min. and 164 α-sec in comparison of that of ^{222}Rn but also are readily adsorbed on the surface of carbon film compared to ^{222}Rn. After 4 days, the density of α-tracks decreased following the decay curve with an apparent half-life of 3.8 days which is in line with the 3.82 days half-life of ^{222}Rn, because excess ^{218}Po and ^{214}Po supported by ^{226}Ra decayed away and are adsorbed away for the first 3 days and the secular equilibrium among ^{222}Rn and its daughters are kept in the solution after 4 days. Therefore, the intersection between the two different decay curves appeared at the third day in the exposure test. This indicates that the density of α-tracks decreases following the decay curve in the secular equilibrium of ^{222}Rn if the initial sources of ^{218}Po and ^{214}Po are not in water. Furthermore, this fact is supported by eq. (7) in our theoretical consideration. We thus could detect the concentration of radon and its daughters in secular equilibrium in water by measuring the density of α-tracks.

(3) Detection of groundwater flowing sections in wells by the aquatic α-track method

Generally, groundwater contains more radon than surface water and rainwater (Fukui 1985). Flowing groundwater contains more radon than stagnant water, because the flowing groundwater can collect fresh radon as it moves through fissures. When we measure the concentration of radon dissolved in groundwater by using the aquatic α-track method, we can obtain a higher density of α-tracks at fissures where groundwater flows faster than that where the flow is stagnant or low. Consequently, we can predict the section of groundwater flow in boring wells on the basis of the profile of density of α-tracks measured in-situ.

We measured the profile of density of α-tracks in wells B-2, B-4, B-5 and B-6 by sinking the aquatic α-track detectors into the groundwater of boring wells for one week. The measured profiles of α-tracks are illustrated in Fig. 5. The density of α-tracks in B-5 and B-6 (of the mountain side) are relatively higher than that in B-4 and B-2 (of the sea side). This suggests that the movement of groundwater at the mountain side is more active than that at the sea side, because the groundwater potential at the mountain side is higher than that at the sea side. Maximum density of α-tracks was found at the depth of EL-5m in the B-5 well. However, the entire profile of density of α-tracks in well B-6 was higher than that in well B-5. Therefore, groundwater movement in B-6 is more active than that in B-5. On the other hand, deep parts (i.e. deeper than EL-50m) of every well were found to have very low density of α-tracks, which suggested that groundwater movement was negligible.

In Fig. 5, we also indicate the characteristic distributions of permeable and impermeable fissures which possibly controls shallow groundwater movement in the site at the vertical section line from B-4 to B-6. Primarily, structural impermeable fissures **A** and **B** are predicted to shut off groundwater moving from B-6 and B-5 to B-2, because the contours of groundwater potential measured as pore water pressure are higher in the B-6 and B-5 side than in the B-2 and B-4 side and the impermeable fissures **A** and **B** play an important role in dividing the groundwater flowing region into two different regions. Namely, groundwater potential in the region of B-5 and B-6 is inversely higher than that in the region of B-2 and B-4. This coincides with the prediction that groundwater in the region of B-5 and B-6 probably flows faster than in the region of B-2 and B-4, and that the region of B-5 and B-6 has a higher density of αtracks than that of B-2 and B-4. On the other hand, highly permeable fissures **C**, **D**, **E** and **F** cross at the depth of EL-25 m to EL- 40 m of the boring well B-2. The highest density of α-tracks in well B-2 is observed at the aforementioned depth where permeable fissures are concentrated. This section in the well is judged to be the paths where groundwater flows. Furthermore, this permeable section was measured as having the highest permeability of the order of 10^{-3} cm/sec.

The distribution of density of αtracks and distribution of permeability do not necessarily agree (in comparison between Fig. 5 and Fig. 6). Inconsistency between the density of α-tracks and permeability is great at deeper parts in each boring well. Furthermore, there is relative good coincidence between the density of fissures and density of α-tracks at shallower parts (less than EL -50 m deep) in each well. On the other hand, there is not coincidence between the density of fissures and density of α-tracks at deeper parts (more than EL-50 m deep) in each well (Fig. 7). These facts indicate that groundwater flows negligibly or very slowly at parts deeper than EL-50 m and that groundwater can not flow unless there is a hydraulic gradient of groundwater even if permeability is great.

We attempted to estimate groundwater ages in order to verify that deep groundwater is older than shallow groundwater. Groundwater apparent age was estimated by the helium accumulation method taking into consideration the groundwater evolution trend on the basis of decrease in the helium isotopes ratio with increase in groundwater ages (Mahara, 1995). Measured helium contents are listed in Table 1 together with sampling depth, neon content and $^3He/^4He$ ratio. The trend of helium isotopes evolution and the correlation between apparent groundwater ages and sampling spots of groundwater are shown in Fig. 8. The oldest groundwater was found at the deep part in well B-4. The youngest groundwater was found at the shallow part in well B-5, and its helium content was similar to that of distilled water equilibrated with atmospheric helium. Groundwater at the deeper part of each well was generally older than at shallower parts. This fact suggests that deep groundwater flows very slowly or is stagnant, while shallow groundwater flows fast. The results estimated from the distribution of density of α-tracks were completely consistent with the evolution trend of groundwater.

Table 1 Contents of ^{4}He and ^{20}Ne, and $^{3}He/^{4}He$ ratios of groundwater samples

No	Well	Depth(m)	^{4}He (ccSTP/g)	^{20}Ne (ccSTP/g)	$^{3}He/^{4}He$
1	S-2	EL-9.49	7.24E-08	2.37E-07	1.22E-06
2	S-2	EL-22.99	7.68E-08	2.34E-07	1.19E-06
3	S-2	EL-31.99	8.45E-08	2.52E-07	1.11E-06
4	S-2	EL-37.99	7.79E-08	2.29E-07	1.13E-06
5	S-4	EL-15.29	1.30E-07	2.88E-07	7.97E-07
6	S-4	EL-61.29	2.05E-07	3.78E-07	5.68E-07
7	S-4	EL-64.29	1.97E-07	3.46E-07	5.89E-07
8	S-4	EL-81.29	1.85E-07	3.03E-07	5.88E-07
9	S-5	EL-0.9	4.42E-08	1.73E-07	1.47E-06
10	S-5	EL-5.1	4.87E-08	2.03E-07	1.40E-06
11	S-5	EL-11.1	4.94E-08	2.00E-07	1.34E-06
12	S-5	EL-28.1	1.04E-07	3.45E-07	1.48E-06
13	S-6	EL-21.37	8.91E-08	2.31E-07	1.02E-06
14	S-6	EL-39.37	9.79E-08	2.23E-07	8.37E-07
15	S-6	EL-64.37	9.58E-08	2.62E-07	9.18E-07
16	S-6	EL-89.37	1.09E-07	2.87E-07	9.25E-06

CONCLUSIONS

We developed the aquatic α-track method to measure dissolved radon and its daughters in groundwater. This method is useful for measuring the in-situ radon dissolved in groundwater without collecting groundwater, which requires the prompt measurement and correcting for the decay loss of radon. We can estimate the groundwater flowing section in the boring well from the profile of density of α tracks measured by the aquatic α-track method. Consequently, radon logging using the aquatic α-track method is useful for detecting groundwater flow paths in groundwater surveys.

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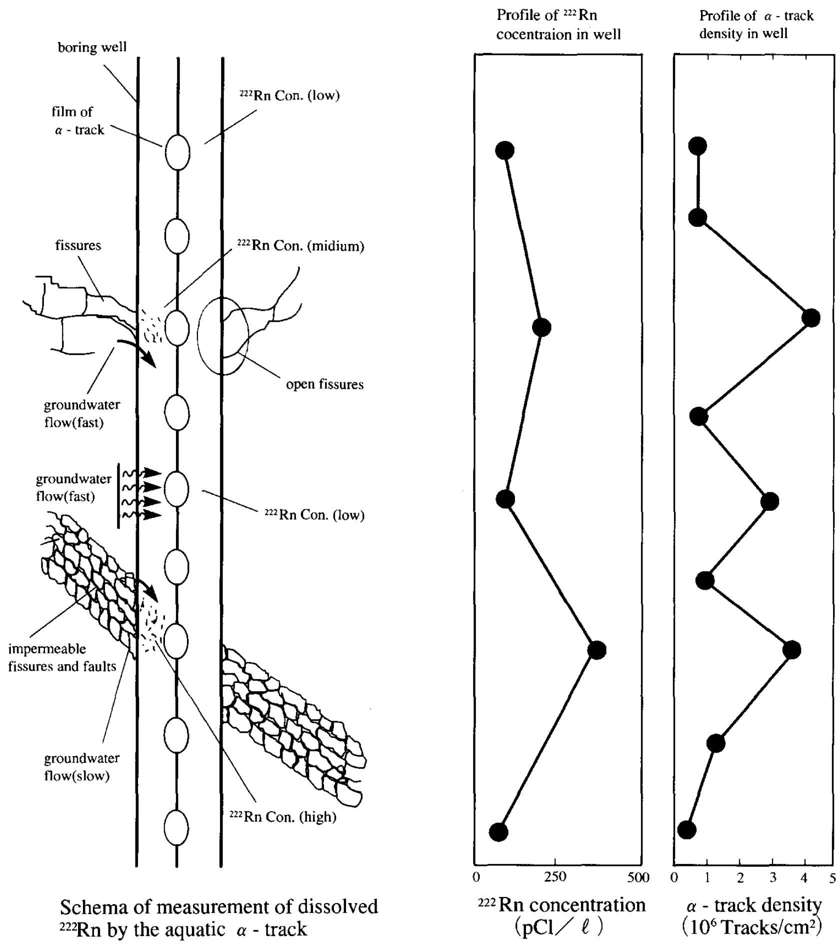

Fig. 1. Schema of the detection of the groundwater flowing section by using the aquatic α -track methrod

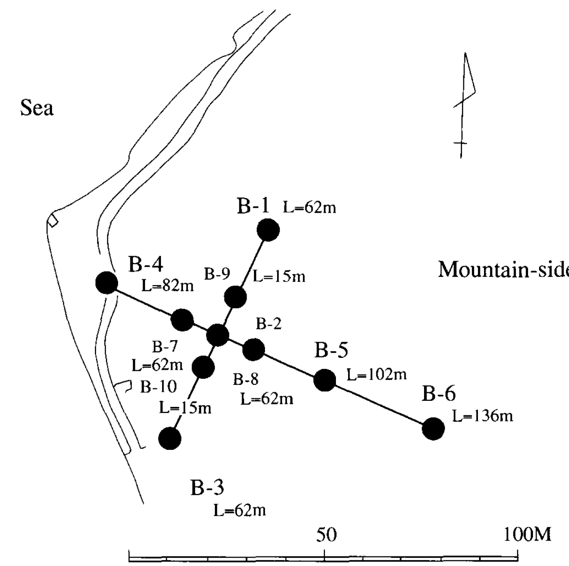

Fig. 2. Location of boring wells in the test site

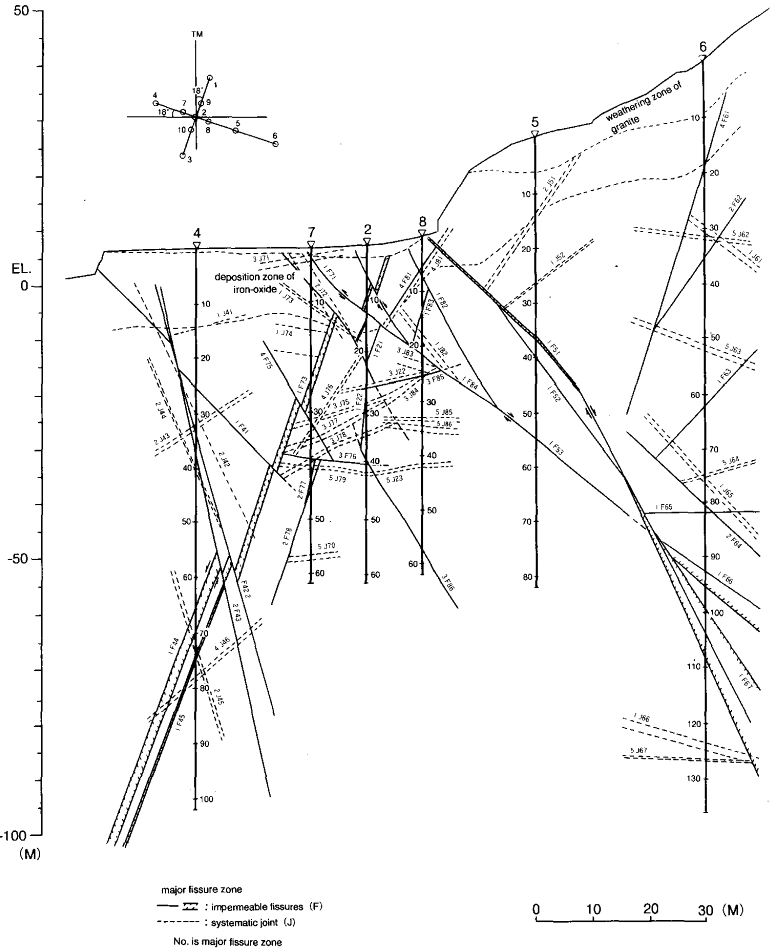

Fig. 3. Distribution of fissures and faults in test site B4—B6 vertical section

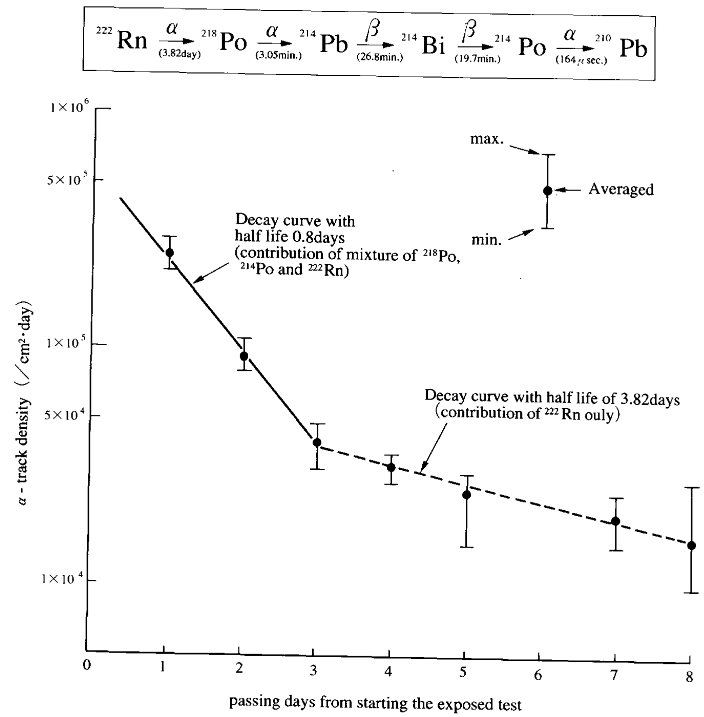

Fig. 4. Relationship between reduction of α - track density measured in water initially equilibrated with ^{222}Rn and the exposed time.

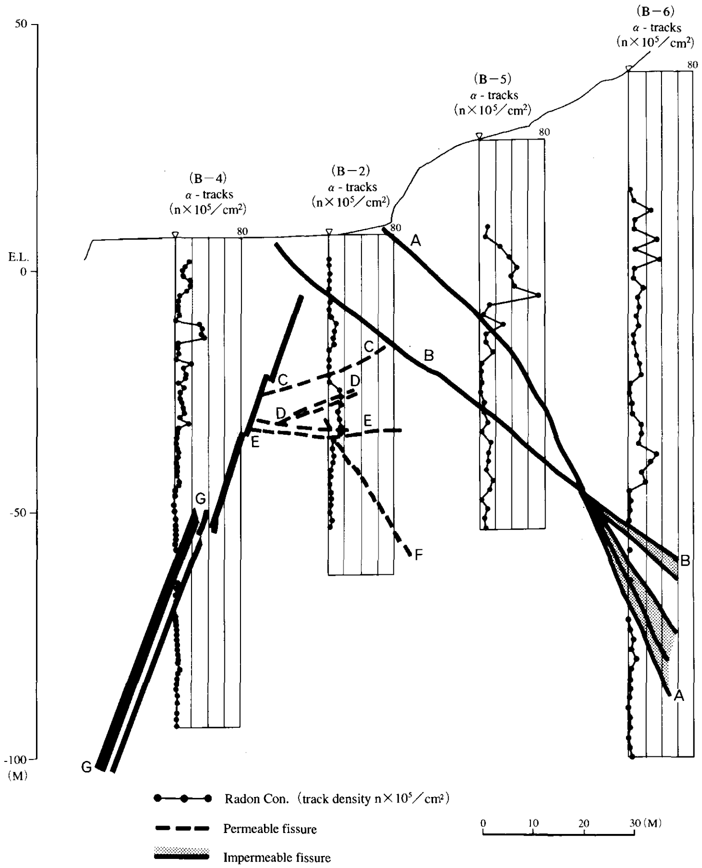

Fig. 5. Correlation between geological distribution of permeable fissures and distribution of α - track density (B4—B6 Vertical Section)

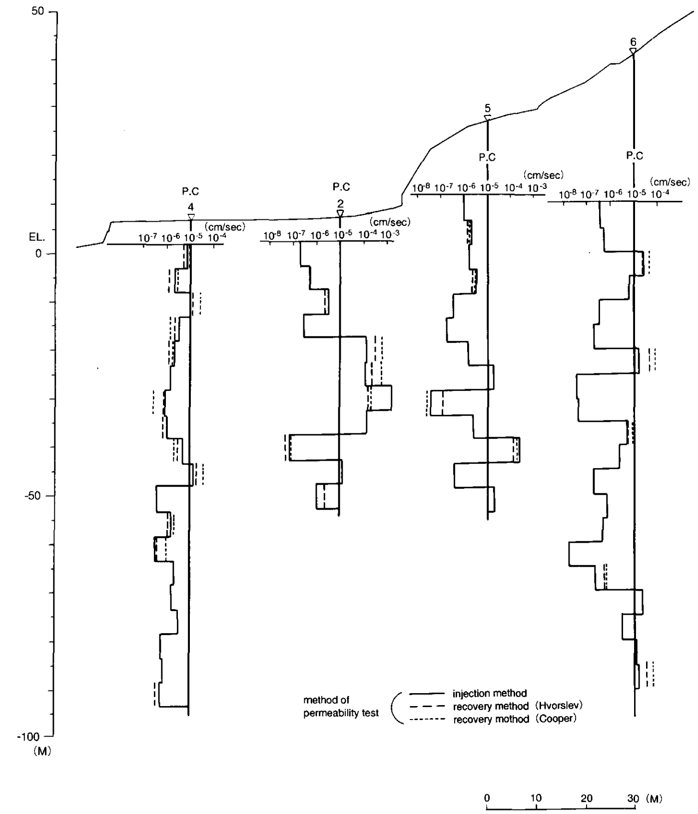

Fig. 6. Distribution of permeability coefficient (P.C) in boring wells

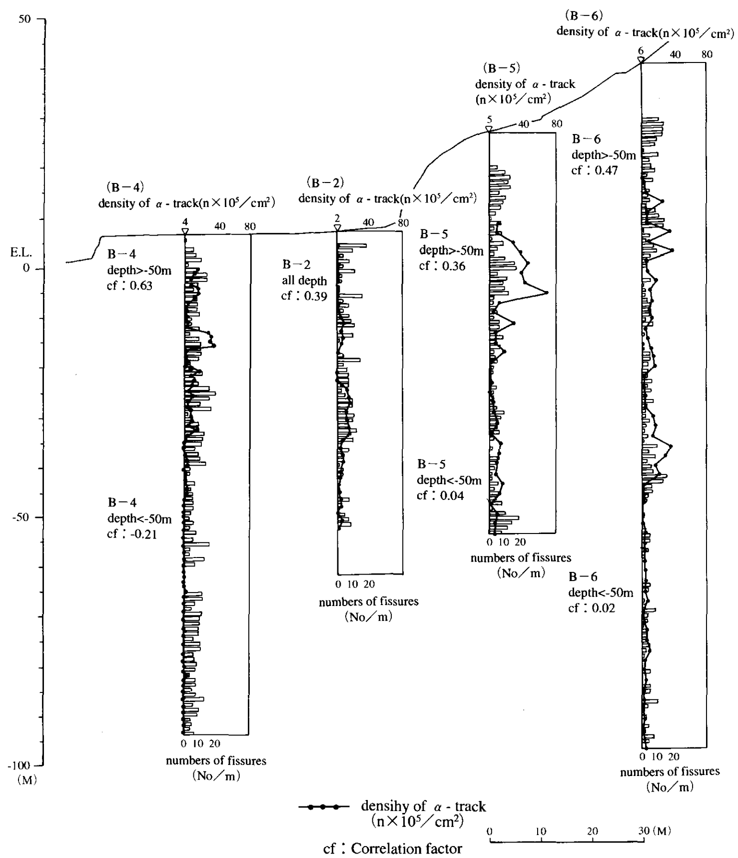

Fig. 7. Correlatrion between the density of fissures and the density of α - tracks (B4−B6 vertical section)

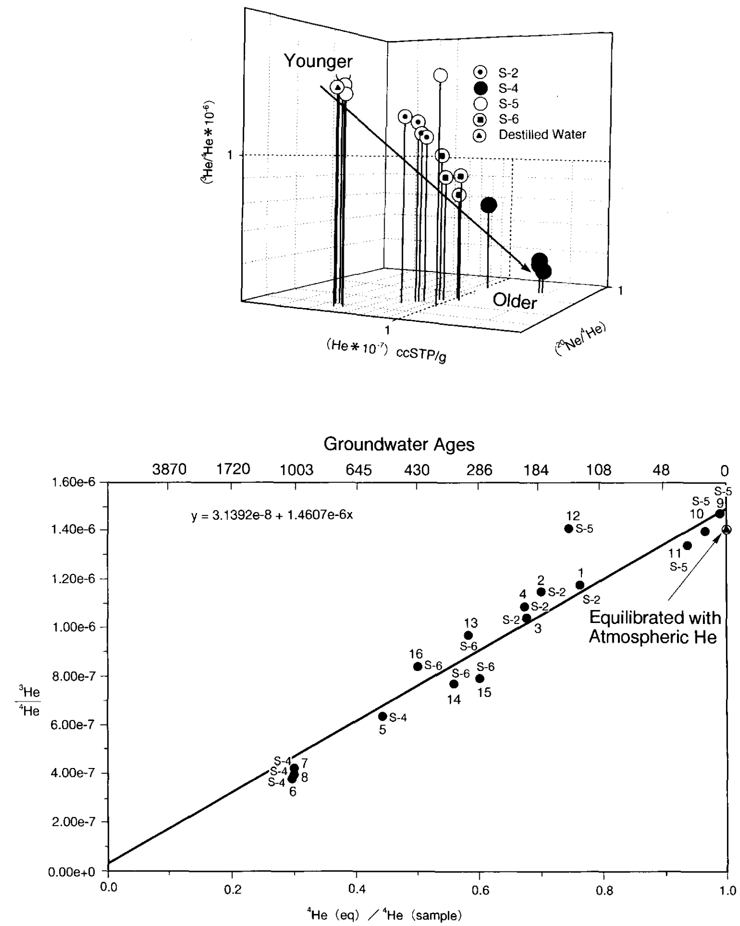

Fig. 8. Estimation of groundwater ages and the groundwater evolution trend from the characteristic changes in the dissolved helium isotopes