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Volcanic Gas Chemistry for Estimating Temporal Changes of Temperature and Pressure in Geothermal Reservoirs

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ABSTRACT

The purpose of this study is to clarify the sensitivity and estimation accuracy of Rn (radon-222) for the change of physical properties, in special temperature and pressure, in geothermal reservoirs. The western side of Aso crater in southwest Japan was selected for a case study site, because two reservoirs were estimated by magnetotelluric survey. During two and half years, Rn concentrations have been measured continuously using the volcanic gases pumped from a borehole with 1 or 1.5-m depth and ionization chamber method. Periodic measurements of Rn and Tn (radon-220) by α scintillation counter method, and six chemical components data (He, H_2, O_2, N_2, CH_4, and CO_2) by gas chromatography have been added at one-week intervals. The Rn concentrations by the periodic measurements at the three sites located in the 2-km range clarified the similarity of general fluctuation patterns, which implies an existence of common factor for reservoir condition changes. Because the continuous Rn data at one site were influenced by the atmospheric temperatures, residual components were obtained by subtracting the trend components from the original data. The chemical component data were used to estimate the temperature and pressure in reservoir: resultant temperature range was 230 to 285 °C and average pressure was 80 MPa. Main factor for producing these changes is probably volcanic seismicity. It was clarified that the residual Rn components correspond generally with the temporal changes of the estimated temperatures and pressures. Average temperatures at the other two sites were estimated as 265 and 304 °C. The differences in the value and changeability of the temperatures may be attributable to source depth of volcanic gases.

1. INTRODUCTION

Estimating temperature and pressure and their temporal changes in geothermal reservoirs is indispensable to geothermal resource exploration, evaluation of reservoir capability, and reservoir maintenance. If these properties are estimative on the ground surface without the use of borehole investigation and well logging, regional geothermal survey with low cost becomes possible by obtaining the data at many points concurrently.

From this viewpoint, we have researched on volcanic gas chemistry in a geothermal area by measuring seven component concentrations, Rn (radon-222), He, H_2, O_2, N_2, CH_4, and CO_2. Among them, major importance was attached to Rn, and two systems for measuring Rn concentrations were adopted: ionization chamber and α scintillation counter methods for continuous and periodical Rn measurements, respectively. Our interest in Rn is based on the pioneer studies by D'Amore *et al*. (1978/79), Cioni *et al*. (1984), Semprini and Kruger (1984), Kraemer (1986), and Kruger and Semprini (1987) which proved usefulness of Rn as an in-situ reservoir tracer. Semprini and Kruger (1984) identified a high correlation between Rn concentration and fluid enthalpy, and proposed a possibility of estimating reservoir temperature from Rn. The usefulness of Rn is attributable to its chemical properties as a noble and inert gas, small solubility in water, and detectable at extremely low concentration. Thus, Rn can remain preferentially with the vapor phase of water in geothermal resources at reservoir temperatures (Kruger and Semprini, 1987).

Those studies use chiefly gases and fluids sampled from wells in geothermal fields for relationship between Rn concentration and thermodynamic properties of reservoirs. Our target is an area with no wells, and temperature and pressure of reservoir are estimated from the above gas components. The purpose of this study is to examine further the sensitivity of Rn for changes of temperature and pressure in geothermal reservoirs. Usefulness of the combination of multiple gas concentrations is demonstrated by clarifying the accordance of general change patterns between the Rn concentrations and the estimated temperatures and pressures.

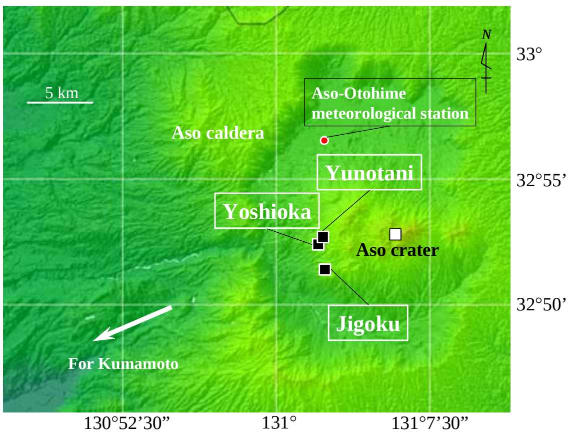

Figure 1: Location of the three hot spring sites for radon-gas measurement in the western side of Aso crater in central Kyushu, southwest Japan.

2. STUDY AREA AND MEASUREMENT SYSTEM

The western side of Aso crater, situated in central Kyushu, southwest Japan, is selected for this case study, because geothermal manifestations in it are clearly seen by the three hot springs, Yunotani, Yoshioka, and Jigoku (Figure 1). Mt. Aso is known to be accompanied by the largest caldera in Japan. These hot springs are located in the 2-km range.

Because the hydrothermal altered zones are extended alonga line connecting the hot springs (NNW-SSE), these are considered to be related to the existence of fracture zone striking this direction.

Because there was no borehole data which reached the geothermal reservoirs, a geophysical prospecting by magnetotelluric survey was carried out at 26 sites around the hot springs concurrently with this study. From the spatial characteristics of resistivity by a 1-D inversion analysis and 3-D interpolation of the resultant resistivities, two reservoirs were estimated in the depth ranges of 2.8-4 km and 1.5-2.5 km (Asaue *et al*., submitted). The Yunotani and Yoshioka sites are located on the deeper reservoir, and the Jigoku site is on the shallower one.

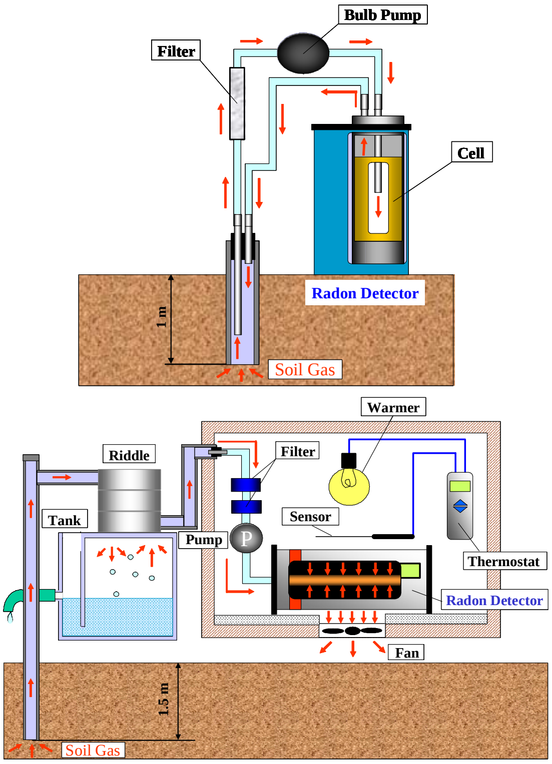

Figure 2: Schematic diagram for two radon measurement systems using α scintillation counter method (top) and ionization chamber method (bottom).

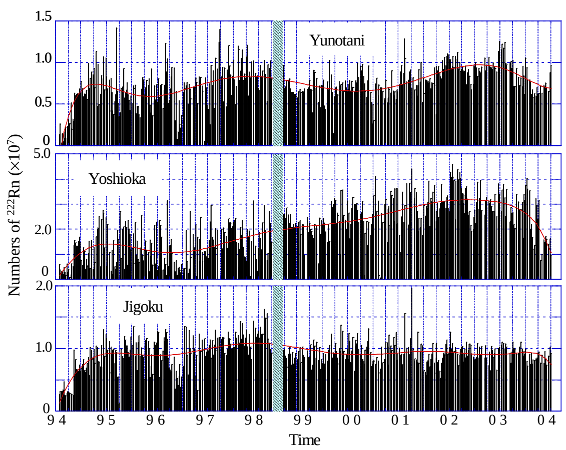

Figure 3: Temporal changes of the numbers of radon, which are calculated from the data by α scintillation counter and the calculation method by Koike *et al.* (2000), at the three hot spring sites.

Six boreholes with 1 or 1.5-m depth and 3-cm diameter (two for each site) were made and protected by a vinyl chloride pipe near the fumaroles in order to collect the volcanic gases. Rn is measured by two different methods: α scintillation counter using RDA-200 detector (Scintrex Inc.) for periodic measurement once a week which started in May 1994 at those three sites; and ionization chamber using Alpha GUARD (Genitron Inc.) for continuous measurement which started in September 2001 at only the Jigoku site. The former method is simple and available for measurements at many sites in short time. Two and half years' data have been successfully accumulated by the latter method. Figure 2 shows schematic diagram of the two methods. The other gas components have been collected at the Jigoku site once a week and quantified by gas chromatography using thermal conductivity detector.

3. TEMPORAL CHANGE OF RN CONCENTRATION

Total numbers of Rn and Tn decays per minute are measured by the α scintillation counter during 20 minutes. The data can be separated into two concentrations, numbers of Rn and Tn at the start of measurement, which are closer to true concentrations than the total decays of Rn and Tn by an empirical equation. Koike *et al*. (1996; 2000) describes this separation method that considers the radioactive equilibrium condition of Rn gas, and proved its effectiveness by case studies.

Figure 3 shows the resultant numbers of Rn at one borehole in each area where almost 10 years data could be obtained. The hatched portion indicates missing data period due to the detector trouble. Polynomial curves which represent general trend in the temporal changes of the data are included in the figure. It is clear that the Rn concentrations are variable with the location: the Yoshioka has the largest concentrations. However, the general fluctuation patterns are similar among the three sites, because the times at which the concentrations take local maximums and minimums are approximately the same. It signifies that the temperature and pressure changes in reservoirs have a common cause regardless of the locations.

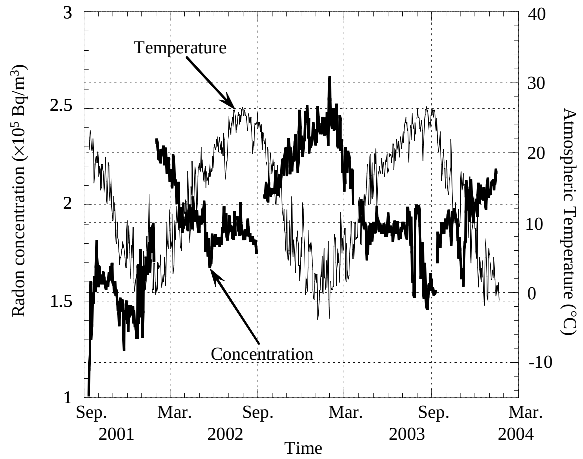

Figure 4: Mean daily radon concentrations at the Jigoku site by ionization chamber method, and its superimposition on the mean daily atmospheric temperatures at the Aso-Otohime meteorological station with 9-km distance (Figure 1).

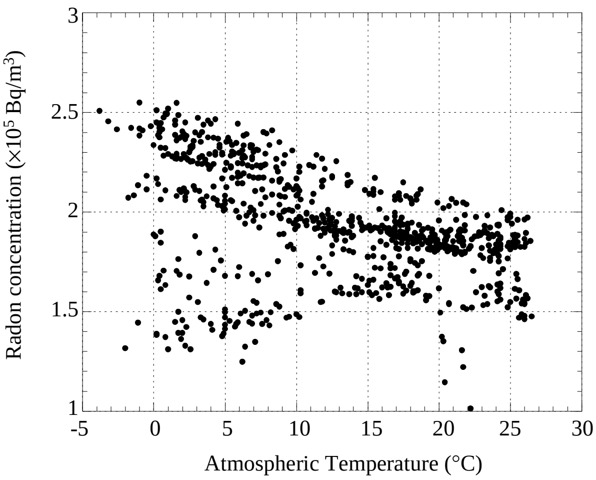

Figure 5: Correlation between the mean daily radon concentrations and the atmospheric temperatures shown in Figure 4.

In addition to the weather effect, a possible main factor for producing the change of Rn concentrations is volcanic seismicity. Volcanic earthquakes with small magnitude frequently occur around the Aso crater. It was found that there are time lags in the response of the change of Rn concentrations to the seismicity, and the lags are variable with the distances to hypocenters.

More detailed characteristics of temporal changes of Rn concentrations are detectable by the continuous measurement. This system measures Rn only with unit of Bq/m^3. Figure 4 shows the mean daily Rn concentrations at the Jigoku site. Four missing data periods were caused by the change of pumps for drawing the gases. Large trend of the Rn concentrations with small fluctuations during short period is clearly seen in the figure. The coefficient of variation is 0.14. Figure 4 also compares the Rn concentrations with the mean daily atmospheric temperatures at the Aso-Otohime meteorological station (Figure 1) whose distance and elevation difference between the Jigoku site are 9 km and 250 m, respectively. This comparison clarifies a negative correlation that the Rn concentrations increase inversely with the temperatures. Such a correlation is weak with the atmospheric pressure or humidity. Because the decay law of radionuclides is not influenced by temperature, this correlation may result from the effect of temperatures on the gas velocity of carrying Rn.

To examine the correlation between the Rn concentrations and the temperatures in detail, a scattergram between them is drawn in Figure 5. This figure shows up that these data cannot be approximated by one regression line because of the existence of several trends. Therefore the Rn concentrations should be divided into several periods to remove accurately the trends related to the atmospheric temperatures. The most suitable time division which generates the highest correlation between the Rn concentrations and temperatures was obtained by the times of changing pump: the total period was divided into five depending on the period for using the same pump. High correlation coefficients between the two data for the five periods were obtained in the range of -0.58 to -0.86, although the total coefficient was -0.45. The difference between the original Rn concentration and the regression line in each period is termed as residual component. Figure 6 shows the resultant residual components that are expected to have relationship with reservoir conditions.

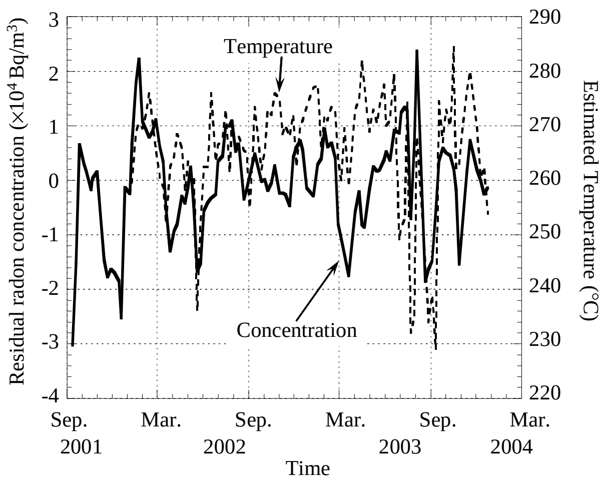

Figure 6: Resultant residual components of the radon concentrations by removing the trends from the original data, and the temperatures estimated from the CO_2, H_2, and CH_4 concentrations.

4. ESTIMATION OF TEMPERATURE AND PRESSURE IN RESERVOIR AND RELATION TO RN CONCENTRATION

4.1 Temperature Estimation

Because there is no data on the temperature and pressure in reservoirs in the study area, these are estimated indirectly from the gas composition. D'Amor (1980) presented multiple equations involving H_2O, CO_2, H_2, CH_4, and H_2S as geothermometers for reservoir temperature, T (K).

Among them, two equations using CO_2, H_2, and CH_4 are:

$$\begin{aligned} &\log(P_{\mathrm{H_2}}/P_{\mathrm{CO_2}}) = 4.34 - 2396/T - \log(Nl_{\mathrm{CO_2}}/Kg_w) \\ &\log(P_{\mathrm{CH_4}}/P_{\mathrm{CO_2}}) = 16.87 - 4318/T - 3.56\log T \\ &\qquad - 1/2\log(Nl_{\mathrm{CO_2}}/Kg_w) \end{aligned} \quad (1)$$

where $(Nl_{\mathrm{CO_2}}/Kg_w)$ is the gas/steam ratio as liters of CO_2 as standard condition per kg of steam, and P means partial pressure. By removing the common term from the two equations, a new equation can be derived as:

$$\log(P_{\mathrm{H_2}}/P_{\mathrm{CO_2}}) - 2\log(P_{\mathrm{CH_4}}/P_{\mathrm{CO_2}}) = -29.4 + 6240/T + 7.12\log T \quad (2)$$

The partial pressures can be replaced by the concentrations of CO_2, H_2, and CH_4.

The above six gas measurements started four months later than the continuous Rn measurement. Figure 6 shows the superimposition of the estimated temperatures using equation (2) for two years and the residual Rn concentrations. Because the gases have been sampled once a week using the same borehole as the Rn measurement, the residuals are weekly averaged. The estimated temperatures vary in the rage of 230 to 285 °C with an average of 265 °C. In almost one year from June 2002 to June 2003, the temperatures are stable with small fluctuations around 270 °C. There may be a correlation between the temperatures and residuals, because the similar temporal changes can be seen without time lags. The noteworthy correspondence is found at the large depressions of temperatures in May 2002 and September 2003: the residual Rn concentrations also largely decrease at these times. Therefore, appropriateness of the removal of trends from the original Rn concentration data and the estimation method of temperature can be confirmed from that correspondence. Figure 7 is another expression for the correlation with a regression line. Although these data are scattered and correlation coefficient is not large (0.49), it is reasonable to conclude that the Rn concentration increases in harmony with the reservoir temperature.

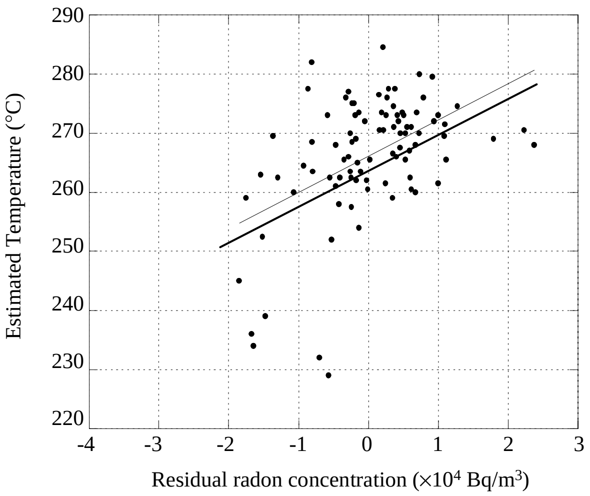

Figure 7: Scattergram between the mean weekly radon residual concentrations and the estimated temperatures shown in Figure 6 with a regression line.

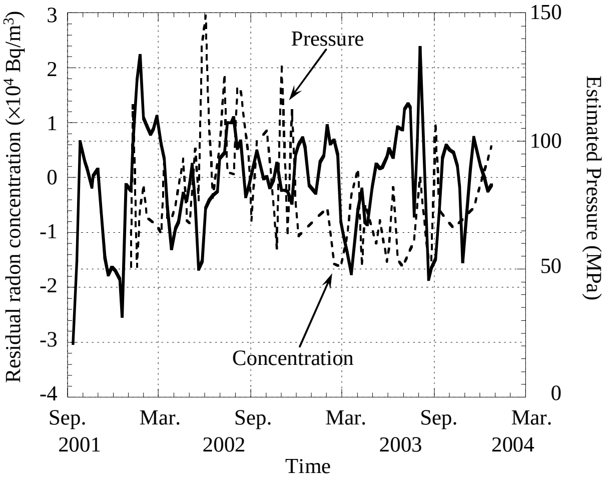

Figure 8: Comparison of the residual radon concentrations and the pressures estimated from the He and CO_2 concentrations.

4.2 Pressure Estimation

Pressure estimation from the gas composition is more difficult than the temperature. Here, we follow the method proposed by Caracausi *et al*. (2003) that the He/CO_2 ratio depends on the pressure of magma degassing. It allows calculating pressures throughout open system by assuming the initial He concentration in vapor within the typical range of mafic magmas (Ozima and Podosek, 1983).

Figure 8 compares the estimated pressures with the residual Rn concentrations. Because this estimation uses only two components less than the temperature estimation, it is more sensitive to the measurement errors. In addition, the average of He concentrations is the smallest (2.0 ppm) among the six components, and some data are missing because of under detection limit. Considering the general pressures calculated, the pressures over 150 MPa are excluded in the figure by regarding them as abnormal values.

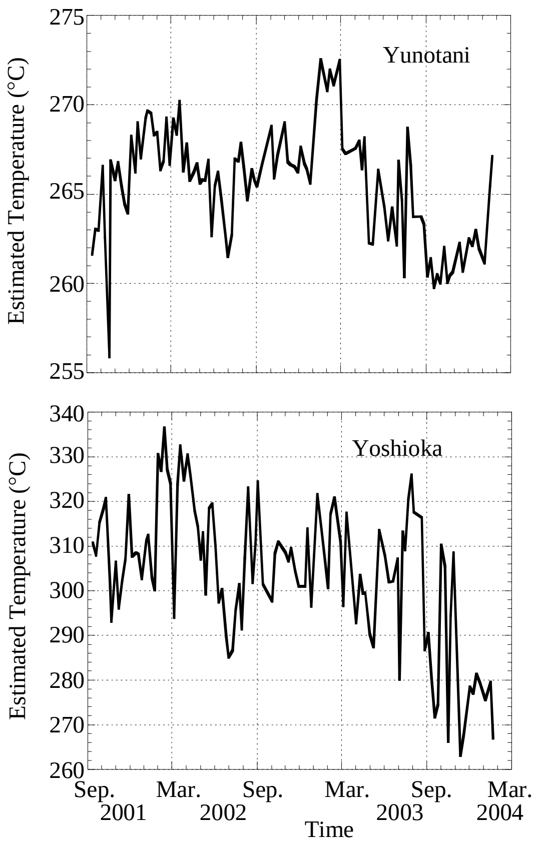

Figure 9: Estimated temperatures after September 2001 at the Yunotani and Yoshioka sites using the data by α scintillation counter method, which is based on the empirical equation between the estimated temperatures and the numbers of radon for the data at the Jigoku site.

Although the pressures are less reliable as compared to the temperatures, they can be used as rough estimates of the reservoir condition. This is because the average pressure, 80 MPa, is not largely different from the lithostatic condition ($\sigma = \rho g h$) at the Jigoku site by assuming rock density (ρ) and depth (h) of the reservoir as 2.7 g/cm^3 and 2.5 km, respectively, which gives σ=66 MPa. The h is based on the result of magnetotelluric survey. The correlation between the pressures and the residual Rn concentrations is low as compared to the estimated temperatures. However, a general trend that the concentrations increase approximately from the start of measurement to August 2002 and decrease toward April 2003 is also appeared in the pressures. It can be considered that the Rn concentration has a positive correlation with both temperature and pressure in reservoirs.

5. APPLICATION OF RN CONCENTRATIONS TO ESTIMATING RESERVOIR TEMPERATURES AT DIFFERENT SITES

At the Jigoku site, the residual Rn concentrations can be compared with the numbers of Rn by α scintillation counter method shown in Figure 3. A positive correlation is found between them, although the correlation coefficient is not high (0.40). Therefore, the empirical equation between the estimated temperatures and the residual Rn concentrations can be extended to an equation using the numbers of Rn as the concentration. This equation is applied to the numbers of Rn at the Yunotani and Yoshioka sites with no gas composition data.

Figure 9 represents the estimated temperatures at these sites after September 2001. Although the distance between the sites is short (0.5 km), the temperatures are higher at the Yoshioka: the averages are 266 °C at the Yunotani and 304 °C at the Yoshioka. Comparing Figures 6 and 9, the times in which the temperatures decrease suddenly such as July 2002 and September 2003 are common among the three sites. It is remarkable that the temperature changes at the Yunotani are small as similar to the Jigoku site: most data are within 260-270 °C. On the other hand, the changes are large within 260-340 °C and the temperatures tend to decrease with the time at the Yoshioka site. Although the Yunotani and Yoshioka sites are estimated to be on the common reservoir by the result of magnetotelluric survey, the source depths of gases may be different: the depth is deeper at the Yoshioka considering the dissimilarity of temperatures and large sensitivities of temperature changes.

6. CONCLUSIONS

For the purpose of estimating indirectly temperature and pressure conditions in reservoirs from volcanic gas chemistry, we examined the capability of Rn and other six components, He, H_2, O_2, N_2, CH_4, and CO_2, at the three sites in the western side of Aso crater, southwestern Japan. The detailed continuous Rn measurement using ionization chamber method, which required a complicated system, was set at only one site. However, the data by simple α scintillation counter method at the other two sites were also available because of a positive correlation between the two kinds of Rn data. Based on the two and half years' continuous Rn data, necessity of removing the atmospheric temperature effect from the data was clarified. The gas compositions were successfully used to obtain estimates of temperatures and pressures. The high capability of Rn as an in-situ tracer for reservoir conditions was demonstrated because the temporal changes of the residual Rn concentrations generally correspond with those of the temperatures, which varied from 230 to 285 °C, and the pressures with an average of 80 MPa.

Although the general trend in the temporal changes of the Rn concentrations was common among the three sites, the average and variance of temperatures at the Yoshioka site were largely high as compared to the Yunotani and Jigoku sites. The average temperatures were 304, 265, and 266 °C, respectively. Because the two sites with different average temperatures were estimated on the same reservoir from the resistivity model by the magnetotelluric survey, the source depths of gases were considered to be different from the dissimilarity and changeability of the temperatures.

The main factor that fundamentally changes temperature and pressure in the reservoirs is probably volcanic seismicity around the Aso crater. The Rn concentrations can fluctuate accompanied by the changes. It can be concluded that our proposed methods are useful to evaluate reservoir physical conditions and identify external factors affecting them.

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