FUMAROLIC RADON EMANATIONS DUE TO ACID ALTERATION IN THE BEPPU GEOTHERMAL SYSTEM, KYUSHU, JAPAN

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SUMMARY - The high-temperature geothermal system of Beppu shows a clear relationship between fluid chemistry and elevation due to the existence of a boiling zone at higher elevations within the 1400 m high Quaternary andesite volcanoes overlooking the city. Although the abundance of radon emanating from the system is highest around fumaroles, the ratios of $^{222}Rn/^{220}Rn$ in steam suggest these isotopes have travelled for only about 10 minutes since leaving their sources. The dominant source of radon in this system, appears to be near-surface acid alteration of andesite by steam condensates rather than deeper sources, such as the degassing of magma. Because previous workers have usually measured *only* ^{222}Rn, acid alteration may have been overlooked as a significant source of radon in other high-tempera —— geothermal systems.

1.0 INTRODUCTION

1.1 Beppu

The city of Beppu, in northeast Kyushu, is one of the most important spa resorts in Japan. According to the City office, 12 million tourists a year are drawn to its spas, with their *onsen* (hot springs used for bathing) and *jigoku* (literally "hells" i.e. fumaroles, steaming ground and boiling springs which are developed as tourist attractions), making it one of the most economically important geothermal systems in Japan.. Approximately 2,800 wells, mostly shallow, but some ranging up to 700 m deep and reaching temperatures of 200 °C, currently produce a total output of hot water and steam of about 1.3 million liters a day, used primarily for spas and space heating. The system has produced about 500 kg/s during more than 50 years of exploitation, with only minimal drawdown *(Allis* and Yusa, 1989). The study reported here focuses on the distribution of radon isotopes in the Beppu area, carried out with the aim of tracing the geothermal circulation, and detecting possible magmatic inputs.

1.2 Geological Setting

Beppu lies at the eastern end of the Beppu-Shimabara Graben (Figure 1). This major structure extends ESE-WSW through north-central Kyushu, in southwestern Japan at the western end of the Median Tectonic Line (Matsumoto, 1979). Within the graben is a negative Bouguer anomaly of up to 40 mgal, which delineates a zone of subsidence covering an area of at least 2000 km^2. Three-dimensional gravity modeling suggests that within the graben near Beppu, pre-Tertiary, granitic basement lies at 1-2 km below sea level (Kowazawa and Kamata, 1985).

The Surface geology of the Beppu area is characterized almost entirely by volcanic rocks, chiefly hornblende andesites (Hoshizumi et al.. 1988). Thus volcanic flows, debris flows and alluvial fans, modified by fault topography, dominate the landscape. Geomorphologically the Beppu area consists of three distinct areas, separated by major faults (Figure 1). To the south the Asamigawa and Yufuin fault systems separate a dissected mountainous area from the graben. To the north is a plateau averaging 700 m above sea level separated from the graben by the Beppu-kita fault system.

The oldest volcanic rock in the area is a propylitized hornblende andesite, believed to be of Pliocene age, overlain by pyroxene-hornblende andesites, rhyolitic pyroclastic flows, and pyroxene andesites, ranging in age from 0.9 to 0.46 Ma. Lavas erupted as the *graben* subsided so that the terrain around Beppu is dominated by the young hornblende andesite volcanoes of Yufudake (1574 m), Tsurumidake (1385 m), Garandake, (1043 m) and Ogiyama (792 m). Tephrochronology suggests these volcanoes began about 35,000 y B.P. with the last eruptions between 1,000 and 2,000 years ago (Kobayashi, 1984). The Beppu Fan, a large alluvial fan, formed by debris flows, covers the eastern flanks of Tsurumidake below 300 m above sea level. This fan underlies most of the city and is the reservoir rock for much of the waterdominated part of the geothermal system.

1.3 Beppu Geothermal System

The Beppu Geothermal System is believed to have originated as a result of this volcanic activity, and the heat sources of the system to lie within the volcanoes Tsurumidake and Garandake. Superheated fumaroles occur near the summits of both volcanoes. Allis and Yusa (1989) estimate that the total natural heat output of the system is at least 250 MWt, and the temperature of the deep reservoir ranges up to 300 °C. Thus the Beppu geothermal system is typical of the many similar large, high-temperature hydrothermal systems around the margins of the Pacific Ocean, hosted in andesite volcanoes and associated with subduction zones.

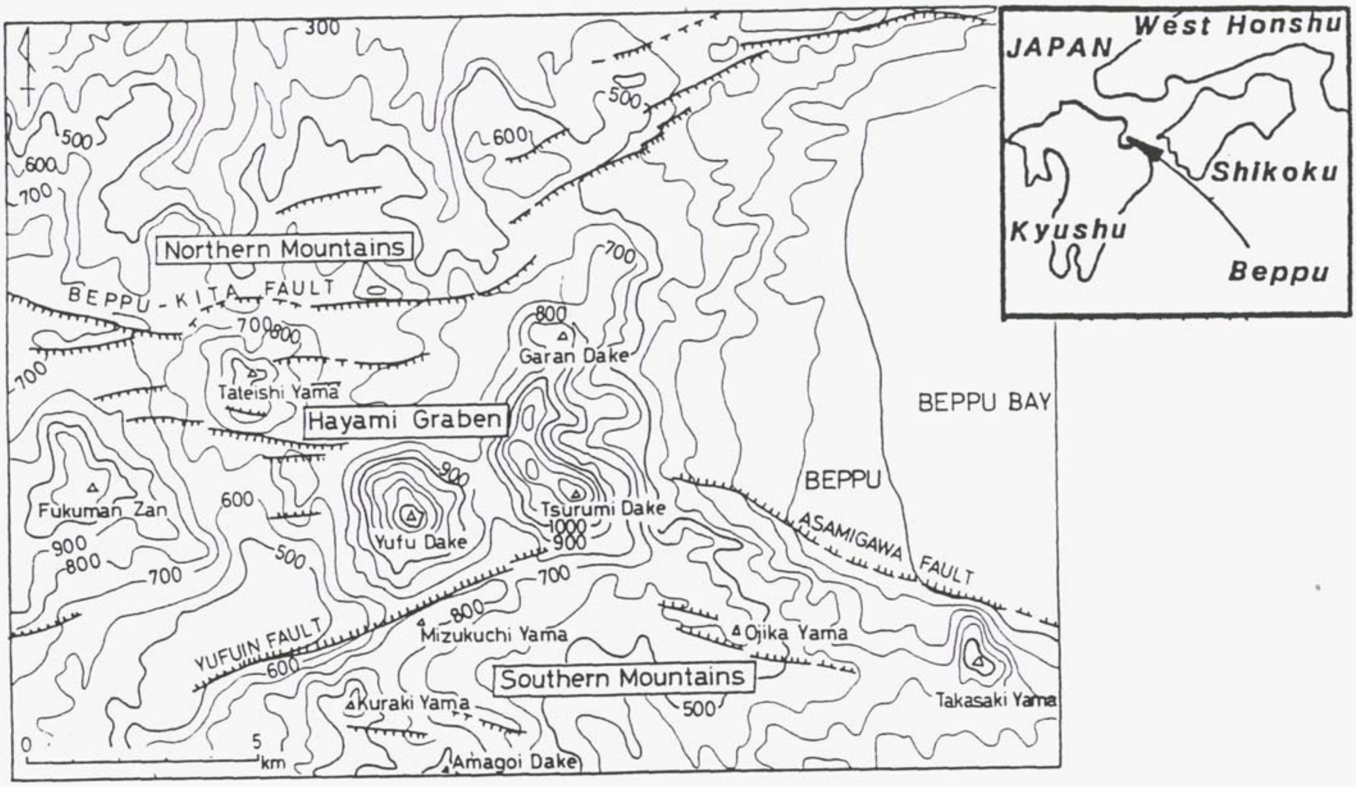

Figure 1- The geomorphological regions of the Beppu Area.
The Hayami Graben forms the eastern end of the Beppu-Shimabara Graben (inset).

The major outflow zones of the system occur at low elevation in the Beppu Fan (Figure 1). These are a southern zone along the Asamigamwa Fault, termed by Allis and Yusa (1989) the "Beppu Thermal Zone", and a northern zone which they termed the "Kamegawa Thermal Zone". There is a clear relationship between water chemistry and altitude, due to the development of a boiling zone at high elevation. Thus the thermal waters of Beppu's spas are noted for their *diverse* chemistry. The principal water types are (1) acidic sulfate water, (2) near-neutral, bicarbonate water, and (3) near-neutral, sodium chloride water. Mixtures of these end members are commonly encountered, together with dilute steam-heated waters, especially in shallow wells (Allis and Yusa, 1989). In general, the sulfate waters (400-600 ppm) are restricted to the western end of the Kamegawa zone at higher elevations, and the bicarbonate waters (<1000 ppm) *occur* in the Beppu thermal zone, especially at its western end. The chloride waters *occur* throughout the remainder of both thermal zones (up to 2000 ppm Cl^- in the Kamegawa zone and 1000 ppm in the Beppu zone). The highest chloride concentrations *occur* at the western ends of the zones, except for where chloride is increased due to an incursion of sea-water at the coastal end of the Beppu zone. A mixed bicarbonate-chloride water predominates in downtown Beppu and a mixed chloride-sulfate-bicarbonate water predominates in downtown Kamegawa.

From these observations and from their interpretation of enthalpy chloride diagrams and chemical geothermometers Allis and Yusa *(1989)* infer the following. (1) The heating zone has a maximum temperature in the range 250-300°C. (2) The geothermal reservoir fluid has a chloride concentration of 1400 ppm and an enthalpy of about 1200 kJ/kg. (3) The chemistry of the Beppu outflow zone is dominated by cooling and dilution of this parent fluid by mixing with shallow groundwater, whereas that of *the* Kamegawa outflow zone is dominated by cooling of the parent fluid by boiling and *steam* loss. (4) Although the thermal area covers an area of about 30 km², the surface manifestations of the system are quite restricted. Furthermore the main discharge zones are displaced 4-5 km laterally from the supposed heat sources. This is due to a combination of the high relief (<1575 m) of the volcanoes and the high typhoon rainfall in the Beppu area.

2.0 RADON STUDIES

2.1 Radon in Geothermal Systems

Radon in hydrothermal systems has been studied in a broad range of contexts including (1) seismicity and volcanic hazards (e.g. Segovia, 1991), (2) exploration for geothermal resources (e.g. Whitehead, 1981), and (3) modeling of transport within geothermal systems (e.g. D'Amore and Sabroux, 1976. Its principal isotopes are ^{222}Rn (half-life 3.823 days), which forms from ^{226}Ra in the ^{238}U decay series, and ^{220}Rn (half-life 54.5 *seconds)*, (also called "thoron") which forms from ^{224}Ra in the ^{232}Th decay series. Many hot springs, fumaroles, in geothermal areas show strong enrichment in radon due to radioactive disequilibria. In such geothermal systems the proportion of radon reaching the *surface* as the result of decay of its parent isotopes in solution is usually much less than that arriving as radon *gas* (Wollenberg, 1975).

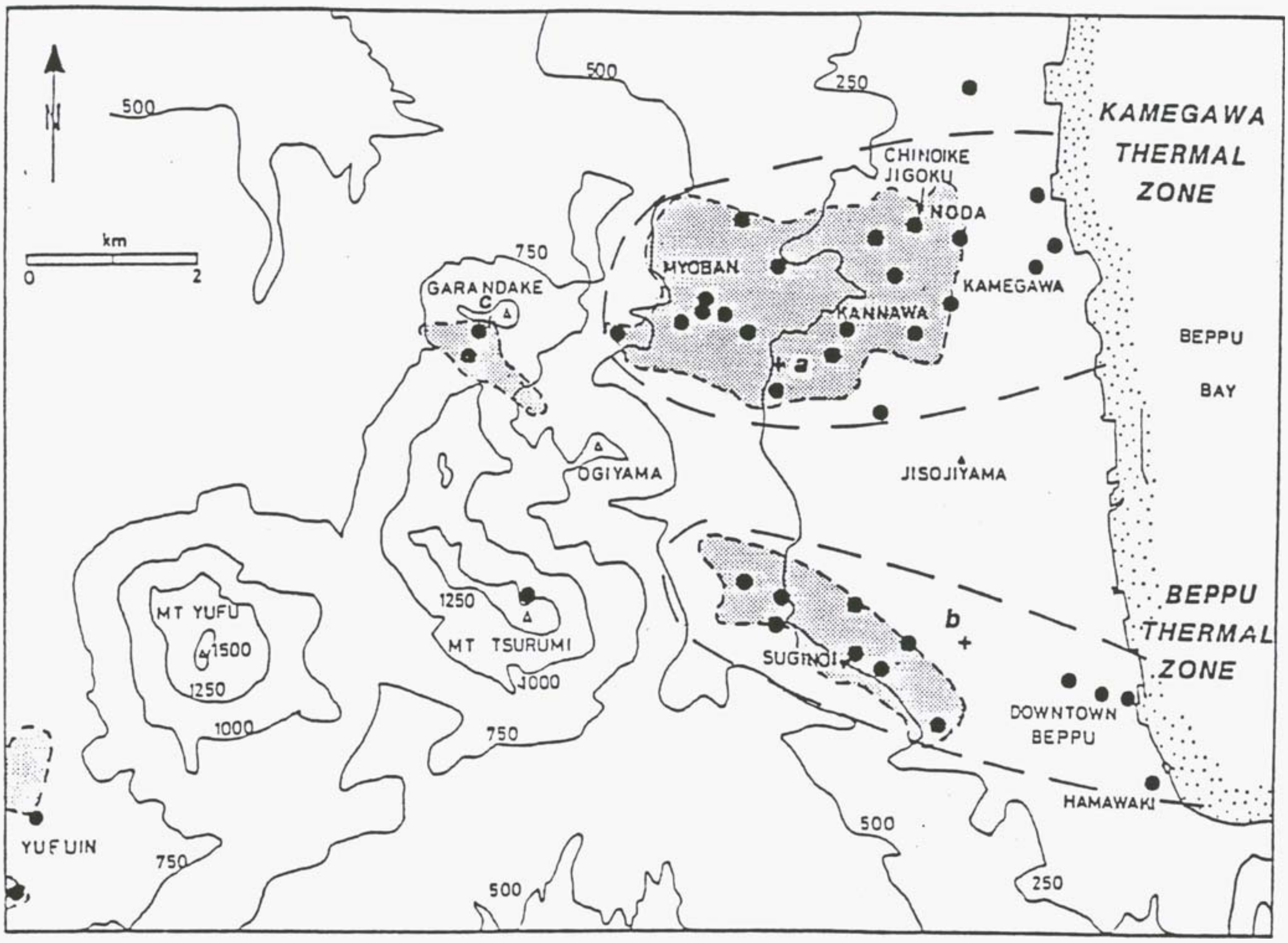

Figure 2-Distribution of natural thermal activity (circles) and hydrothermal alteration (stippled). (Modified from Allis & Yusa, 1989, Fig.3)

Such anomalously high radon outputs are usually regarded as either being due to magmatic degassing or to localized zones of rapid upwelling of *steam* as, when boiling occurs, radon, a noble *gas*, fractionates to the *steam* phase.

2.2 Previous Radon Studies at Beppu

Koga *et al.* (1957) studied the concentration of radon and radium in waters from 50 hot springs and radium in 11 hot spring deposits from Beppu, using a liquid scintillation counter for separated radon *gas* and an ionization counter to *analyze* radium precipitated as $RaSO_4$. They found that the radioactivity of the Beppu geothermal system is low. The thermal waters analyzed contained $< 0.1 \times 10^{-9}$ nCi/l (curies/liter) of radon, with the highest value being 0.52 nCi/l. Hot springs in Japan classified as radioactive contain > 30 nCi/l. Half of the Beppu water samples had radium contents of 0.1×10^{-12}g/l (pg/l) with the highest value being *6.5* pg/l. In the hot spring deposits typical concentrations of radium were < 0.5 parts per trillion (ppt) with the highest value being 10.7 ppt. These concentrations are about the same as those of typical andesites around Beppu which contain 0.3-0.5 ppt of radium. Their data also showed that only *5%* of the radon in the thermal waters could be attributed to the decay of radium in solution. Thus 95% must be transported as *gas* (*Koga et al.*, 1957).

In 1983 Koga returned to his *study* of radon in the Beppu geothermal system. He sampled *steam* from boiling springs and *steam* vents from 18 sites in the Kamegawa Thermal Zone and 11 sites in the Beppu Thermal Zone. He analyzed the non-condensible fraction for radon using a scintillation counter. Because the time elapsed between collection and sampling time was thought to be too long for ^{220}Rn to Survive in measurable quantities, Koga (1983) reported the results as ^{222}Rn. His data indicated that in the Beppu Geothermal Zone the concentration of radon in *steam* is low, averaging only 1.22 nCi/l, with the highest value being 4.44nCi/l. In the Kannewa Zone they are also low, averaging 1.1 nCi/l in the eastern part, and 4.59 nCi/l in western part. However in the center of this zone they average 5.3 nCi/l, with a high value of 11.3 nCi/l. Koga (1983) concluded, that because of the low concentration of radon, firm decisions on the significance of the data could not be drawn. However he suggested that the higher radon flux occurring in the central part of the Kannewa Zone, was due to it having higher permeability and closer proximity to the heat source for the geothermal system.

3 0 NEW STUDIES OF RADON

31 Aims

The purpose of this study was to re-examine the utility of radon isotopic data in understanding the hydrothermal system at Beppu, extending the study to soil gases as well as to thermal waters, and analyzing $^{222}Rn/^{220}Rn$ ratios to test models of distant versus local sources of the gas.

32 Soil Gas Results.

Our initial survey measured ^{222}Rn and ^{220}Rn in soil gases in several hundred sites spread over a wide area around Beppu between sea-level and the summits of the volcanoes. Measurements, obtained by pumping the soil gas into a portable scintillation counter immediately after driving a 1 m deep hole into the ground, were compared with those obtained using alpha track detecting films emplaced in the 1 m deep holes for periods of up to several days (Elders *et al.*, 1992). The data on radon in soil gases fiom the two methods correlate reasonably well and confirm that emanations of radon around Beppu in general are quite low, especially on the Beppu fan at elevations below the zone of boiling, and even at the summit and southern slopes of Tsurumidake. Given this low background level of radon, it was disturbing to find that radon emanations in soil gas at control sites at the Beppu Geophysical Research Laboratory show significant daily variations of a factor of three or four in alpha count rate. These variations do not appear to be related in any simple way with meteorological effects (Elders et *al.*, *1992).*

Furthermore, over the whole area variations in these low-level radon emanations in soil gas did not correlate with type of bed-rock, subsurface temperature, hydrology, or water chemistry, nor were they obviously related to major faults or other structures. However in the vicinity of fumaroles and acid-altered steaming ground the levels of radon outputs in soil gas are orders of magnitude higher. The highest radon emanations correlated with high ground temperatures and zones of acid alteration (Elders et *al*, 1992). It was decided therefore to focus on the origin of these high levels of radon emanations, and to compare them with the ratio of $^{222}Rn/^{220}Rn$, measured by a scintillation counter (EDA RD-200), in steam from fumaroles, Steaming ground., and boiling geothermal wells. This report focuses on those results and their significance.

3.3 $^{222}Rn/^{220}Rn$ Ratios and Transit Times.

Measuring the ratio of $^{222}Rn/^{220}Rn$ permits limits to be put on the transit time of radon gas from its source if the two following reasonable assumptions are valid. (1) At time t_o radon isotopes emanate from source areas in secular equilibrium with their ultimate parents, ^{238}U and ^{232}Th, with an initial ratio of $^{222}Rn/^{220}Rn$ of R_o, given by, $R_o = {}^{222}N_o/{}^{220}N_o$, where N_o is the number of atoms present when t = 0. We further assume that the most likely value of R_o = 0.4, as this is the average value of the atomic ratios of U/Th for Quaternary andesites in the Beppu area (Ando et al., *1987). (2)* Steady-state "pipe-line" flow occurs, i.e., radon flows fkom the source area to the collector subject only to decay without addition or fractionation. After a travel time of "t" seconds, $R_t = {}^{222}N_t/{}^{220}N_t$, or

$$R_t = {}^{222}N_o e^{-\lambda 222 t}/{}^{220}N_o e^{-\lambda 220 t} \quad (1)$$

where λ refers to is the decay constants of the respective isotopes. Hence:-

$$R_t = R_o e^{(\lambda_{220} - \lambda_{222})t} \quad (2)$$

By monitoring the decay in alpha radiation of a radon sample over successive time periods (usually 60 s) with the scintillation counter, the ratio R_t can be determined. At the collector the count rate for α decay is proportional to λN_t. Thus C_t ,the ratio of the count rates for the two isotopes at time t is :-

$$C_t = C_{222}/C_{220} = (\lambda_{222}/\lambda_{220})({}^{222}N_t/{}^{220}N_t) \quad (3)$$

Combining equations (1) and (2), the relation between C_t and t is given by:-

$$C_t = (\lambda_{222}/\lambda_{220})R_o^{(\lambda_{220} - \lambda_{222})t} \quad (4)$$

We obtain $(\lambda_{222}-\lambda_{220})$ and $(\lambda_{220}/\lambda_{222})$ from the half lives of the isotopes concerned. Table 1 shows the count rate ratio for various possible transit times for situations where the *two* assumptions above are valid.

4. RESULTS

41 Sampling Radon in Steam

A rapid sampling method was developed to determine the ratio of radon isotopes in *steam.* This allowed *steam* to pass from the collection system into a condenser cooled by a dry ice/ethanol bath, and the non-condensible fraction to be introduced into the cell of a scintillation counter within one half-life of ^{220}Rn. Experiments showed that the results were reproducible for multiple samples from the same source, even when taken on different days. Collection sites included high, medium and low elevations in both the Kannewa and Beppu Thermal Areas, fumaroles, vents and steaming ground, boiling wells spanning a range of water types.

Table 1 Relation between Transit Times and Count Ratios

t (seconds)	$C_t = C_{t222}/C_{t220})$
0	6.58×10^{-5}
1	6.66×10^{-5}
10	1.47×10^{-5}
100	2.35×10^{-4}
1000	22.0

Table 2. Radon Output in Typical Steam Samples (Counts per minute). Count Ratio (C_t) and Calculated Transit Times

Type	Remarks	C_{222} (cpm)	C_t	t (sec.)
Neutral Chloride Wells	Low elevations 100-200 m	75-140	0.05-0.4	522-690
Boiling Pools	Elevation 250- 350 m	152-195	0.07-0.36	548-668
Mixed Water Wells	Elevation 350-400 m	190-615	0.17-0.35	617-630
Vents and Steaming Ground	Highly altered 375 m asl	615	0.35	634
Superheated Well and Vents	Highly altered 650 m asl	40-930	0.21-0.47	634-697
Steam vents	Highly altered 1300 m asl	420-2300	0.46-0.07	695
Fumarole (135°C)	Highly altered 1000 m asl	740-750	0.47	695

4.2 Radon Data for Steam

Radon was determined in *steam,* sampled as described above, from approximately 35 sites at elevations between 100 and 1300 m elevation, from both vapordominated and waterdominated zones and including both the Kannewa and Beppu Thermal Areas. The sites sampled included superheated (< 135°C) and non-superheated fumaroles, vents and steaming ground, boiling pools, and superheated and non-superheated boiling wells, with various types of thermal waters (Elders, et al., 1992). Typical results are shown in Table 2, together with the tranit times calculated fkom these data. As can be seen fkom the Table, in all of the samples analysed the counts per minute for ^{220}Rn far exceed those for ^{222}Rn, with $^{222}Rn/^{220}Rn$ ratios (C_t) in the range 0.05 to 0.47. Steam samples from neutral chloride waters from boiling wells at low elevations, in both the Kannegawa and Beppu Thermal Areas, exhibit the lowest radon outputs and lowest C_t values. Steam from boiling pools and mixed bicarbonate-sulfate-chloride waters have higher, but more variable, radon outputs.

The highest radon outputs are from steam vents in areas of steaming ground in *areas* with extensive hydrothermal alteration at moderate to high elevation., Steam fkom such vents and steaming ground has the highest total radon outputs of up to 2,300 cpm for C_{222} and 7,000 cpm for C_{220}, with C_t as low as 0.07.

5.0 SUMMARY AND CONCLUSIONS

Our work on the Beppu Geothermal System, extending the work of Koga (1983) to a larger area and a wider variety of sample types, confirms that, in general, radon output is relatively low. The lowest total radon in *steam* is found in boiling wells with neutral chloride water. Higher outputs are found from vigorously flowing and superheated fumaroles. However superheated fumaroles at high elevation on the volcano, which reasonably might be the most likely candidates for a "pipe model" with magmatic input of radon. are not the sites of the highest radon and thoron output. The highest outputs are found from vents in areas of acid alteration.

Our data further indicate that sampling and analysis of thoron is possible with reproducible results. In all of our samples of soil gas and steam output of thoron exceeds that of ^{222}Rn. Assuming that the ultimate sources of radon and thoron are ^{238}U and ^{232}Th, respectively, and that radon and thoron move in secular equilibrium, *steam* samples with the lowest thoron to radon ratio should be the furthest travelled from their source. We note that these are also the steam samples with the lowest radon output. so that the radon in the neutral chloride waters is either fractionated or the most distant from its source.

The $^{222}Rn/^{220}Rn$ ratios allow calculation of *the* transit times froman andesitic magma or source rock for a "pipe model". Although the relationship between C_t and t is not particularly sensitive, all these calculated transit times lie in the range of 500-700 seconds, or 9 to 13 half-lives of thoron. If radon is being *emitted* from a magma chamber at a depth of 4.5 km say, then these transit times imply unreasonably high velocities in the range of *6-9* ms^{-1} (23-32 km/h) for the *gas* to reach the *surface.* It *seems* particularly significant that the highest radon outputs and the highest thoron to radon ratios *occur* in areas which also exhibit the highest degree of acid alteration. These observations and the short transit times in tum imply that the sources are local, and that the most efficient radon and thoron source in the Beppu Geothermal System is acid alteration by *steam* condensate of andesitic rocks near the surface. Because previous workers usually measured only thoron, acid alteration may have been overlooked as a significant source of radon in other high-temperature geothermal systems.

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