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NATURAL RADIOACTIVITY IN GEOTHERMAL WATERS, ALHAMBRA HOT SPRINGS AND NEARBY AREAS, JEFFERSON COUNTY, MONTANA

By Robert B. Leonard and Victor J. Janzer

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DISCLAIMER

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FACTORS FOR CONVERTING ENGLISH UNITS TO METRIC UNITS

The following factors can be used to convert English units used in this report to equivalent metric units.

Multiply	By	To obtain
feet (ft)	0.3048	meters (m)
gallons per minute (gal/min)	.06309	liters per second (L/s)
inches (in)	25.40	millimeters (mm)
miles (mi)	1.609	kilometers (km)
pounds (lb)	.4536	kilograms (kg)

NATURAL RADIOACTIVITY IN GEOTHERMAL WATERS, ALHAMBRA HOT SPRINGS AND NEARBY AREAS, JEFFERSON COUNTY, MONTANA

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ABSTRACT

Radioactive hot springs issue from a fault zone in crystalline rock of the Boulder batholith at Alhambra, Jefferson County, in southwestern Montana. The discharge contains high concentrations of radon, and the gross activity and the concentration of radium-226 exceed maximum levels recommended by the Environmental Protection Agency for drinking water. Part of the discharge is diverted for space heating, bathing, and domestic use.

The radioactive thermal waters at measured temperatures of about 60°C are of the sodium bicarbonate type and saturated with respect to calcium carbonate. Radium-226 in the rock and on fractured surfaces or coprecipitated with calcium carbonate probably is the principal source of radon that is dissolved in the thermal water and discharged with other gases from some wells and springs. Local surface water and shallow ground water are of the calcium bicarbonate type and exhibit low background radioactivity. The temperature, percent sodium, and radioactivity of mixed waters adjacent to the fault zone increase with depth.

Samples from most of the major hot springs in southwestern Montana have been analyzed for gross alpha and beta. The high level of radioactivity at Alhambra appears to be related to leaching of radioactive material from fractured siliceous veins by ascending thermal waters, and is not a normal characteristic of hot springs issuing from fractured crystalline rock in Montana.

INTRODUCTION

The purpose of this report is to describe anomalously high radioactivity in thermal waters in Alhambra Hot Springs, located about 12 miles (19 km) southeast of Helena (fig. 1), and to suggest the relation of the hot springs to the local and regional geology and hydrology.

Several studies in the United States and elsewhere have described higher levels of radioactivity in thermal waters than in adjacent cooler ground water and surface water (Scott and Barker, 1962; O'Connell and Kaufmann, 1976). Radioactive decay may be a major source of the geothermal heat (Tilling and Gottfried, 1969; Blackwell and Robertson, 1973; Gabelman,

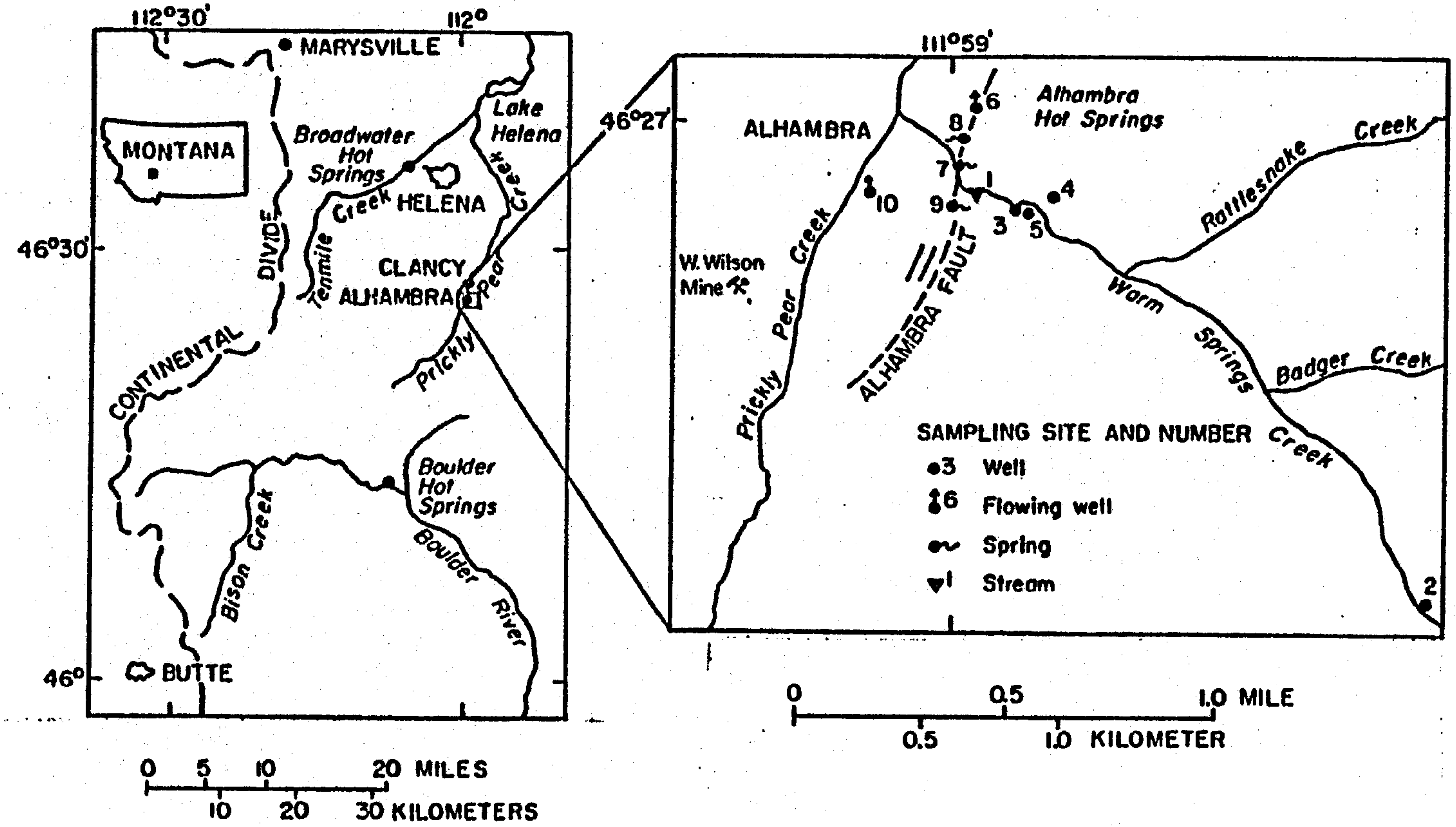

Figure 1.--Index map showing location of wells and springs in the Alhambra area.

1976). Radiochemical studies by Wollenberg (1975), Stoker and Kruger (1975) and others suggest that radiochemical data can be a useful tool for the exploration and evaluation of geothermal resources. Moreover, the potentially deleterious effects of elevated levels of radioactivity on public health (U.S. Environmental Protection Agency, 1976) could impose severe restrictions on the development of geothermal resources. Data describing radioactivity of thermal waters are thus essential to an assessment of Montana's geothermal resources.

During a reconnaissance survey of local uraniferous deposits with a field-type Geiger counter, Thurlow and Reyner (1952) detected about twice the normal background levels of radioactivity at Alhambra Hot Springs and Boulder Hot Springs, about 20 miles (32 km) to the south (fig. 1). They reported that carbonate incrustations around the Alhambra springs registered more than 10 times normal radioactivity background and that radioactivity was detected with a scintillometer while traversing the area by automobile. During a reconnaissance by the USGS in 1970, slightly higher than normal levels of radioactivity at Alhambra Hot Springs and normal background levels at Boulder Hot Springs were recorded with field equipment (E. C. Robertson, written commun., 1976). At both localities the thermal waters are used for space heating, bathing, and domestic purposes. Therefore, anomalously high levels of radioactivity could constitute potential hazards to health.

The cooperation and assistance of local landowners, particularly Mrs. W. L. Bomparte, Mr. R. C. Butler, and Mr. W. W. Wall, and of Mr. W. J. Lindsay, a drilling contractor of Clancy, are sincerely appreciated. Mr. Larry Lloyd, health physicist of the Montana Department of Health and Environmental Sciences, deserves special recognition for his continued advice and support, including the release of unpublished data included in this report. His contributions to the investigation do not necessarily imply total agreement with the conclusions.

GEOHYDROLOGIC SETTING

Three groups of hot springs, several zones of seepage, and a flowing hot-water well lie along a 0.3 mile (0.5 km) segment of the Alhambra fault (Smedes, 1966) in the valley of Warm Springs Creek near its mouth (fig. 1). Thermal waters issue from fractured quartz monzonite and alaskite or from the shallow overlying alluvium and colluvium at a combined rate of about 250 gal/min (16 L/s). Mean annual precipitation of about 20 inches (500 mm), chiefly in the form of snow in the Elkhorn Mountains to the east, sustains perennial streamflow and provides recharge to the fractured rock aquifers.

The hydrothermal system sustaining the Alhambra hot springs probably is confined to igneous intrusive and volcanic rock. Alhambra is located in the northeast part of the Boulder batholith, a composite pluton of Laramide (Late Cretaceous to early Tertiary) age that is composed primarily of quartz monzonite (Smedes, 1966; Knopf, 1963). The monzonite is typically sheeted and locally is crossed by pronounced systems of jointing. Dikes, plugs, and irregular masses of aplite, alaskite, and younger rhyolite (age 35.8±1.4 m.y., R. A. Chadwick, Montana State University, written commun., 1976) are typically shattered and sheared.

Nearly vertical silicified zones, locally called "reefs," transect the quartz monzonite and the alaskite bodies. The reefs consist mainly of brecciated country rock containing many small discontinuous lenses of cryptocrystalline silica, some of which contains highly radioactive uranium minerals (Roberts and Gude, 1953). Although the reefs are resistant to erosion, forming prominent ridges and spurs, they have been faulted and somewhat brecciated before and after silicification that occurred soon after emplacement of the batholith (Pinckney, 1965).

Radioactivity in the batholith appears to be associated preferentially with siliceous rock. Samples of silicic Butte quartz monzonite and alaskites similar to the rock exposed near Alhambra Hot Springs contain 5.9 and 9.2 ppm (parts per million) uranium; 22.3 and 36.3 ppm thorium; and 3.6 and 4.5 percent potassium, respectively (Tilling and Gottfried, 1969, table 2). Average concentrations for the Boulder batholith are lower.

Near Alhambra, primary and secondary uranium minerals are concentrated in fissure fillings of microcrystalline quartz and chalcedony associated with siliceous reefs. Roberts and Gude (1953) reported that a sample of high grade ore from the abandoned W. Wilson mine, about 0.6 mile (1 km) southwest of the hot springs, assayed 9.58 percent uranium. The main reef in which the mine is located trends about N.60 E. (Smedes, 1966). If continuous beneath alluvial and colluvial deposits, the reef would intersect the Alhambra fault in the subsurface near the hot springs.

The crystalline rocks, including the reefs, presumably are too old to represent sources of residual heat, and no firm evidence is available to support contact of the thermal waters with younger hot rock. Radioactive elements in the rocks of the Boulder batholith are sufficiently abundant to produce a significant part of the average measured heat flow of about 2 microcalories per square centimeter by radioactive decay (Blackwell and Robertson, 1973). However, decay of radioactive elements in localized deposits probably is a negligible source of heat for the hot springs.

The mean annual air temperature at Alhambra is about 6°C. If geothermal gradients of 0.64°C/100 feet (21°C/km) measured in similar rock about 5 miles (8 km) to the southwest (Blackwell and Robertson, 1973) can be extrapolated to Alhambra, the maximum temperature of 59.4°C recorded for the hot springs theoretically would be expected at a depth of 8,200 to 9,800 feet (2.5 to 3 km). Evidence from a geothermal test well at Marysville and the deep mines at Butte (fig. 1) indicates that faults and fractures similar to those observed at Alhambra act as conduits for deep circulation of meteoric waters through rock that normally is impermeable when intact.

METHODS OF INVESTIGATION

Samples for radiochemical analysis were collected by the USGS at hot spring 9, flowing hot well 6, and Boulder Hot Springs (fig. 1) to substantiate the previously reported anomalies. Samples were also collected at Broadwater Hot Springs, about 2 miles (3.2 km) west of Helena, because the springs represent a somewhat similar geohydrologic environment and are under consideration for development as a source of space heating and domestic water supply. Only the samples from Alhambra showed a level of radioactivity above normal background. There, the gross alpha and beta activity and the concentration of radium-226 exceeded levels recommended for drinking water (U.S. Environmental Protection Agency, 1976).

Mr. Larry Lloyd, Montana Department of Health and Environmental Sciences (MDHES), and the senior author made a reconnaissance survey during the summer of 1976 using an SPA3 scintillometer[1] to localize the sources of radioactivity at Alhambra. Observed levels of radioactivity were 5 times normal background near the hot springs and seeps and as much as 10 times normal background in travertine deposits around one of the vents and in siliceous zones in adjacent fractured crystalline rock. A similar scintillometer survey of the Broadwater Hot Springs failed to reveal abnormal levels of radioactivity. Analyses by MDHES for gross alpha and gross beta activity of selected water samples confirmed abnormal levels of radioactivity at the Alhambra Hot Springs and normal background levels at Boulder and Broadwater.

Additional samples for determination of dissolved uranium, radium-226, and radon by the National Water Quality Laboratory of the USGS in Denver were then collected mainly at sites where the MDHES analyses showed abnormal levels of radioactivity. Some duplicate samples were also sent to MDHES for concurrent analysis.

SAMPLING PROCEDURES

At springs having multiple vents, the samples were obtained where the specific conductance, discharge, and temperature were highest. All the waters sampled were clear and virtually free of suspended matter.

Unfiltered samples for analysis by the USGS were collected in 4-liter polyethylene bottles and acidified with reagent grade hydrochloric acid to a pH less than 1. Unfiltered samples for radiochemical analysis by the MDHES were collected in 1-liter polyethylene bottles and acidified with reagent grade nitric acid to a pH of less than 1. The analyses for gross alpha and beta normally were completed within 1 week of collection.

Samples of thermal waters for determination of dissolved radon-222 were collected in 50 mL (milliliter) heavy-walled borosilicate glass collection

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[1] Use of trade names in this report is for descriptive purposes only and does not constitute endorsement by the U.S. Geological Survey.

tubes to which lengths of Tygon plastic tubing were attached. The lower (short) length of tubing was bent in the form of a U so that the open end was about 1.6 feet (0.5 m) above the sampler. When the desired sampling depth was reached, the trapped air was released through the longer length of tubing by opening a pinchcock at the surface. When a representative sample had been collected by alternately raising and lowering the sampler, the pinchcock was closed and the plastic tubes at each end were sealed for shipment. To minimize decay of the radon-222 (half-life, 3.823 days), the samples were shipped to the laboratory by priority mail for immediate processing.

Duplicate samples, filtered and unfiltered, normally were collected for chemical analysis using methods approved by the USGS, with some modifications suggested for thermal-spring samples by Presser and Barnes (1974). Routine field measurements included water discharge using a Hoff or pygmy current meter, temperature using a maximum-reading mercury thermometer and a thermistor-wheatstone bridge combination, pH using a Sargent-Welch PBL pH meter, specific conductance using a Yellow Springs Model 33 SCT meter, and alkalinity by titration. At several sites, samples of gas were collected in plastic and glass syringes. These samples were analyzed for nitrogen, oxygen plus argon, carbon dioxide, and methane by gas chromatography in the USGS laboratories in Menlo Park, California.

LABORATORY PROCEDURES

Detailed descriptions of the radiochemical procedures used by the USGS for water and sediment analysis are given by Thatcher, Janzer, and Edwards (1977). Determination of radon dissolved in water or mixed with gas or air is made by an alpha scintillation technique, which is similar to the technique for determining radium-226 using radon.

Determination of the gross alpha and gross beta-gamma radioactivity of a water sample is relatively simple and rapid. The results are useful as an indication of total sample radioactivity or as a screening technique. Analyses for dissolved uranium, radium-226, and radon-222 by the USGS were made only on those samples for which the gross alpha activity as determined by the MDHES was greater than 10 pCi/L (picocuries per liter).

LABORATORY RESULTS

The values of gross alpha and gross beta reported for duplicate samples by the USGS generally are much higher than corresponding values reported by the MDHES, particularly for samples containing high levels of gross alpha activity (table 1). Despite the apparent discrepancies, both laboratories verified anomalously high levels of radioactivity at the same sites, and the results varied somewhat proportionately from site to site. The results of recent proficiency tests conducted by the Environmental Protection Agency (EPA) confirm the comparability of MDHES gross alpha data with reference standards distributed by EPA and suggest that the determinations of gross

Table 1.—Analyses of thermal and nonthermal waters

Sample location number (fig.1)	Source	Location		Date of collection	Specific conductance (μmhos/cm^3) at 25°C	Temperature (°C)	Dissolved solids (sum of constituents) (mg/L)	Sodium-adsorption ratio	Dissolved silica (SiO_2) (mg/L)	Gross alpha[1]		Dissolved natural uranium (μg/L)	Dissolved radium-226 (pCi/L)	Dissolved radon-222 (pCi/L)	Potassium-40 (pCi/L)	Gross beta[2]	
		Latitude	Longitude							USGS (μg/L)	MDHES (pCi/L)					USGS (pCi/L)	MDHES (pCi/L)
1	Warm Springs Creek.	46 26 51	111 58 48	6/29/76	-	-	-	-	1	-	5	-	-	-	-	-	2
				8/20/76	192	16.0	130	0.3	21	-	3	-	-	-	-	-	<3
2	Cold well---	46 25 54	111 57 15	8/20/76	384	10.0	251	1.0	14	-	12	-	-	-	-	-	10
3	Cold well---	46 26 49	111 58 44	6/29/76	295	8.9	227	1.0	24	-	3	-	-	-	-	-	6
				10/19/76	-	-	-	-	-	-	4	-	-	-	-	-	<6
4	Cold well---	46 26 52	111 58 36	6/29/76	410	16.1	334	1.7	29	-	11	-	-	-	-	-	18
				10/19/76	-	-	-	-	-	-	12	-	-	-	-	-	8
5	Cold well---	46 26 48	111 58 41	8/03/76	1,000	17.7	632	6.7	48	26	14	7.2	3.5	-	6.3	19	22
				10/13/76	-	-	-	-	-	-	6	-	-	-	-	-	10
6	Flowing hot well.	46 27 01	111 58 50	4/29/76	1,040	52.2	613	11	58	410	-	-	28	-	7.4	110	-
				6/29/76	1,080	54.0	-	-	61	240	112	.9	27	11,000[3]	7.1	69	56
				12/02/76	1,000	52.2	-	11	-	360	110	.8	28	24,000	7.2	90	70
7	Hot spring--	46 26 53	111 58 51	6/29/76	1,110	56.7	651	12	60	-	97	-	-	6,000	-	-	48
				10/19/76	-	-	-	-	-	-	70	-	-	-	-	-	54
8	Hot spring--	46 26 59	111 58 50	6/15/76	1,070	57.7	-	-	-	-	68	-	-	-	-	-	54
				6/29/76	827	59.4	909	12	67	290	94	.9	40	3,000	7.5	81	48
				10/19/76	-	-	-	-	-	-	81	-	-	-	-	-	43
9	Hot spring--	46 26 47	111 58 58	4/28/76	1,580	54.4	953	14	61	930	-	-	72	10,000	13	220	-
				6/15/76	1,600	55.6	-	-	-	-	153	-	-	-	-	-	131
				6/29/76	1,610	56.1	-	-	-	-	214	-	55-70	7,000[4]	-	-	100
				12/02/76	1,510	54.4	-	14	-	890	293	<.4	73	17,000	12	210	150
10	Flowing warm well.	46 26 52	111 59 14	7/10/76	1,680	27.0	1,060	15	67	-	77	-	-	-	-	-	59
				7/13/76	1,650	29.0	-	-	-	260	-	.5	27	-	15	81	-
				7/23/76	-	-	-	-	-	-	113	-	-	-	-	-	60
				10/19/76	-	-	-	-	-	-	121	-	-	-	-	-	53
				12/02/76	1,620	30.0	-	15	-	560	113	.8	37	37,000	15	130	77

[1] Theoretical conversion for natural uranium: 1μg/L = 0.68 pCi/L

[2] As cesium-137

[3] Corresponding gas sample 130,000 pCi/L

[4] Corresponding gas sample 80,000 pCi/L

beta may be slightly lower than the actual values (Larry Lloyd, oral commun., 1977).

To be useful, comparative measurements of duplicate samples should be reported in the same units or be readily convertible to equivalent units. The USGS gross alpha measurements in water are reported in terms of micrograms per liter (μg/L); that is, mass units of equivalent natural uranium (Un) per liter, in preference to picocuries per liter, the activity units used by the MDHES and others. A conversion factor of 1 μg/L = 0.68 pCi/L is used to compare gross alpha determinations in this report.

A curie is the unit of radioactivity equivalent to 3.7×10^{10} disintegrations per second. A picocurie (pCi), or 1×10^{-12} curie, yields 2.22 disintegrations per minute and is one of the more common units used to describe the low level of radioactivity generally associated with environmental samples. Some confusion and uncertainty in reporting gross alpha measurements in water have resulted from the definition of a "special curie" for natural uranium and thorium, which was made by the National Committee of Radiation Protection (Ney, 1974; Hursch, 1974). Use of the "special curie" results in a conversion of mass to activity for natural uranium of: 1 μg = 0.33 pCi. If the three isotopes U-238, U-235, and U-234 found in natural uranium in the proportions 99.28, 0.711 and 0.0006 percent, respectively (Edwards, 1968), occur in the same proportions in the standard isotope calibration solutions, the conversion 1 μg = 0.68 pCi is valid. Perhaps coincidentally, use of the "special curie" conversion factor of 0.33 would result in closer agreement between the results of the USGS and MDHES for high concentrations of gross alpha.

Natural uranium is commonly recommended and used as the gross alpha calibration isotope (American Public Health Association and others, 1976; American Society for Testing and Materials, 1976). Strontium-90 or cesium-137 is recommended as the gross beta calibration isotope. Natural uranium for alpha, and cesium-137 for beta reportedly are the calibration standards of reference for both laboratories. Therefore, the results should agree if the calibration solutions used are made from reliable standards preferably calibrated by or traceable to the National Bureau of Standards. Perhaps "natural" uranium salts used as standards for the gross alpha determinations should be tested to verify the coexistence of the isotopes in their natural abundance.

The maximum contaminant levels for public water supplies specified by the Environmental Protection Agency (1976) for gross alpha and gross beta radioactivity are 15 and 50 pCi/L, respectively. The 50 pCi/L level for gross beta applies if the average annual concentrations of tritium and strontium-90 are less than 20,000 and 8 pCi/L, respectively. Levels of these isotopes in the ground waters from the Alhambra area could reasonably be expected to be less than 500 and 0.1 pCi/L, respectively.

Gross alpha levels determined by the USGS ranged from 26 μg/L (18 pCi/L) in the sample from cold well 5 to 930 μg/L (630 pCi/L) from hot spring 9 (table 1). The corresponding determination by the MDHES for cold well 5 was 14 pCi/L; the maximum determined was 293 pCi/L (compared with 890 µg/L = 605 pCi/L reported by the USGS) for hot spring 9. Gross beta values ranged from 19 pCi/L in the sample from cold well 5 to 220 pCi/L (as cesium-137) from hot spring 9.

Although the MDHES analyses show lower levels of radioactivity than the USGS analyses for corresponding samples, both show that the samples of thermal water from wells and springs 6-10 exceed the maximum contaminant level for gross alpha and beta activity. According to the USGS analysis, gross alpha in the sample from cold well 5 exceeded the maximum contaminant level, but the MDHES determination was lower than the maximum contaminant level.

Radium-226 concentrations ranged from 3.5 pCi/L in cold well 5 to 73 pCi/L in hot spring 9. The drinking water regulations (U.S. Environmental Protection Agency, 1976) state that the maximum combined contaminant level for radium-226 plus radium-228 in community water systems is 5 pCi/L. With the exception of cold well 5, all the samples analyzed by the USGS contained concentrations of radium-226 alone exceeding the maximum contaminant level for radium.

None of the samples were analyzed for radium-228, a beta-emitting radium daughter resulting from the alpha decay of thorium-232. However, the predominance of thorium in the rocks suggests that radium-228 is present in the thermal waters. Activity ratios Ra-228/Ra-226 larger than unity have been reported from several surface- and ground-water sources in Colorado (Johnson, 1971); therefore, the concentration of radium in the waters may be significantly higher than reported.

Maximum concentration of radon-222 was 37,000 pCi/L in a water sample collected from flowing warm well 10. The reported levels probably are 5 to 10 percent lower than the true levels as a result of losses by diffusion of radon through the plastic tube closures. All-glass sampling tubes would have been preferable; however, these were not used because of their fragility, expense, and unavailability at the time of sample collection. Drinking water regulations of the U.S. Environmental Protection Agency (1976) do not specify a maximum contaminant level for radon.

The reported concentrations of potassium-40, a beta-emitter, and dissolved uranium are relatively insignificant from a public health standpoint. For example, the maximum concentration of potassium-40 was 15 pCi/L from flowing warm well 10. Based on an estimated potassium content of the human body of about 0.35 percent (Lange, 1969) and a specific activity of potassium-40 in natural potassium of about 0.75 pCi/mg (picocurie of beta particles per milligram), about 200,000 pCi of potassium-40 is present in a human weighing 165 lb (75 kg). Similarly, the reported concentrations of natural uranium, with the possible exception of the value of 7.2 µg/L for cold well 5, represent relatively insignificant contributions to the gross alpha content of the waters. The higher concentration of uranium in cold well 5 than in associated hot springs resembles similar occurrences in Nevada (Wollenberg, 1975).

SOURCES OF RADIOACTIVITY IN WATER

Either natural or manmade radioactivity, or both, are found in varying concentrations in all precipitation, and surface and ground waters. The natural radioactivity is caused by traces of: 1) the long-lived parent members of three natural radioactive decay series; U-238 (uranium series), U-235 (actinium series), and Th-232 (thorium series) and their daughters; 2) single long-lived radioactive elements, with potassium-40 being the most abundant; and 3) radionuclides such as tritium and carbon-14, which are produced by cosmic rays as well as by nuclear devices.

Uranium occurs in low concentrations in many natural waters; average sea water contains about 3 μg/L dissolved uranium (Gabelman, 1976). U-238 is the most abundant (99.28 percent) natural isotope of uranium. Its decay products, U-234, Th-230, Ra-226, Rn-222, Po-218, Po-214, and Po-210 decay by alpha emission. In terms of their activities (decay rates), U-238 and U-234 are about equally abundant in nature. The average activity ratio U-234/U-238 of about 1.4 in geothermal waters is suggested by a recent study in the western United States (O'Connell and Kaufmann, 1976). U-235 is much less abundant in nature (normally 0.7 percent of natural uranium) than U-238 and does not contribute significantly to the radioactivity of natural waters.

Although thorium is a major radioactive constituent of acid igneous rock, measured levels of thorium-232 greater than a few nanograms per liter (10^{-9} g/L) are rarely found in surface or ground waters. Thorium has been found in detectable concentrations in mine drainage from acid igneous rocks (V. J. Janzer, 1977, unpublished data) but because of the apparent immobility of the ions under normal conditions, thorium generally is not a significant dissolved radioactive constituent of natural waters. However, the beta-emitting daughter radium-228 may, when combined with radium-226, occur at concentrations approaching the maximum contaminant level of 5 pCi/L established by the U.S. Environmental Protection Agency (1976).

RELATIONSHIP OF RADIOACTIVITY TO MAJOR DISSOLVED CONSTITUENTS

As water passes through the soil and rocks it dissolves varying quantities of the minerals, both radioactive and non-radioactive, with which it comes in contact. Therefore, the chemical composition of the water reflects the composition of the rocks as well as variations in temperature, pressure, and travel time along its path in the subsurface.

All the waters sampled near Alhambra were of the calcium bicarbonate or sodium bicarbonate type. The water in general progresses from calcium bicarbonate type streamflow (No. 1) and shallow ground water (wells 2-5) to more highly radioactive sodium bicarbonate type thermal water (wells and springs 6-10) with increasing temperature, concentration of dissolved solids and silica, and sodium-adsorption ratio (fig. 2, table 1). The sodium-adsorption ratio (U.S. Salinity Laboratory Staff, 1954) is defined as:

$$\frac{Na}{\sqrt{\frac{Ca+Mg}{2}}}$$

with all concentrations in milliequivalents per liter. It is normally used for describing the quality of irrigation waters but is convenient for describing differences in the cation distribution in thermal waters and, at Alhambra, for distinguishing thermal waters (high ratio) from the normal ground water (low ratio).

The relatively dilute calcium bicarbonate water from cold well 2 about 1.5 miles (2.4 km) upstream from the hot springs (fig. 1) is probably characteristic of the nonthermal shallow water in the fractured quartz monzonite. The radioactivity is near or below the level of detection, as it is in chemically similar waters from Warm Springs Creek and cold well 3 (table 1), respectively. Levels of radioactivity significantly higher than normal background, but far lower than in the thermal waters, were reported for selected samples of waters in wells penetrating similar aquifers several kilometers from the hot springs (Larry Lloyd, written commun., 1976).

The thermal waters from the Alhambra fault zone (Nos. 6-9, fig. 1) are remarkably similar chemically to the cooler, but slightly more mineralized, discharge of flowing warm well 10. Although less radioactive than the discharge from hot spring 9, which the well was meant to replace, the well water is too radioactive for domestic use. The similarities suggest a common source.

Discharges from cold wells 3-5 are mixtures of thermal and nonthermal waters. The temperature of the water in cold well 5 increased with depth. If the well were drilled deeper, the percentage of thermal water and the radioactivity in the discharge probably would be higher.

Calcium (in equivalent weights) is more abundant than sodium in most of the crystalline rock that underlies the Warm Springs watershed (Smedes, 1966); therefore, the predominance of calcium in the surface water and cool shallow ground water is not unexpected. The relatively low proportion of calcium and magnesium in the thermal waters, as indicated by the high sodium-adsorption ratio (fig. 2) probably results mainly from the decreasing solubility of these carbonates and the increased solubility of sodium salts with increasing temperature. Despite the relatively low concentrations of calcium, solution-mineral equilibrium calculations (Mariner and others, 1976) show that hot spring 8 was saturated with calcium carbonate. Active deposition of calcium carbonate as radioactive travertine at the vent indicates saturation of hot spring 9 also.

According to Thurlow and Reyner (1952) a sample of the travertine or of a similar incrustation at a vent north of Warm Springs Creek contained 0.058 percent equivalent uranium, but only 0.001 percent actual uranium and 10 times normal radioactive background. The high values of radioactivity in

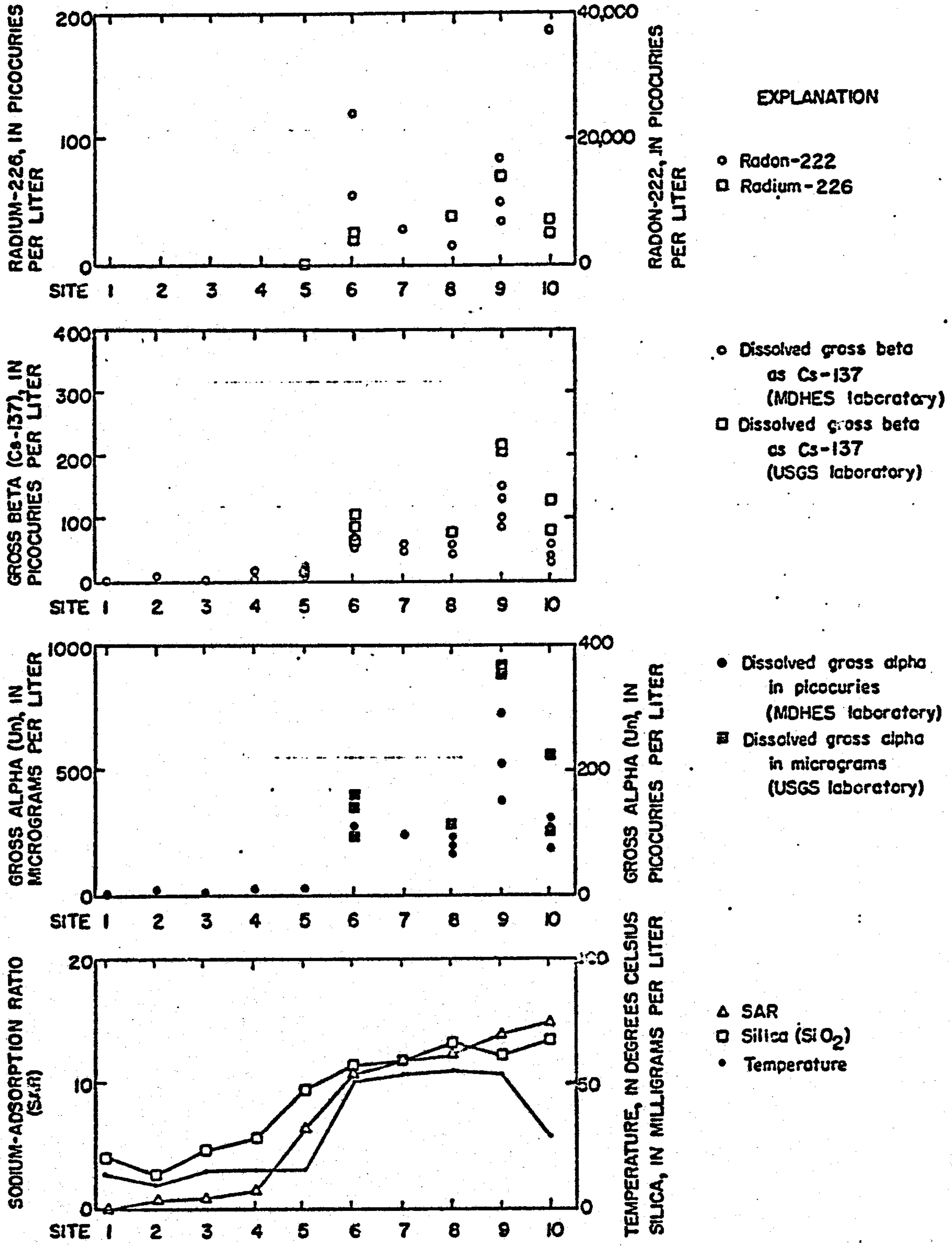

Figure 2.--Variations in radioactivity, sodium-adsorption ratio, temperature, and concentration of silica in water samples.

the discharge and in the travertine are consistent with Wollenberg's (1975) observation that high levels of radioactivity in Nevada thermal waters are commonly associated with calcium-carbonate dominated, in contrast to silica-dominated, hot springs. Most of the major hot springs in southwest Montana have been sampled by the USGS and analyzed for gross alpha and beta by MDHES. Besides Alhambra, only three other springs show high levels of radioactivity and all three are actively depositing calcium carbonate; none of the three issue from fractured crystalline rock.

The relatively insignificant concentrations of dissolved uranium in the waters at Alhambra suggest that radium-226 and its short-lived alpha-emitting polonium daughters (Po-218 and Po-214) are the principal sources of the high alpha activity. High concentrations of radon-222 from several sources confirm the presence of its parent, radium-226, in the system (table 1).

High concentrations of radon are commonly found in water from fractured zones in crystalline rock. The fracturing that accompanies seismic activity presumably increases the surface area of radioactive rock from which radon can be released and transferred to the water. Alhambra lies along the northern Intermountain Seismic Belt (Smith and Sbar, 1974) and is subject to continuing seismic activity that may maintain open fractures in the rock. The epicenter of three major earthquakes, one in 1925 and two in 1935, lies within 20 miles (32 km) of Alhambra (Friedline and Smith, 1974; Robertson and others, 1976).

Much higher concentrations of radon were found at the same sites in December 1976 than in April or June 1976. The concentrations of radon were not proportional to the concentration of radium-226 in the samples nor were fluctuations in the two constituents correlative (fig. 2). Radium-226 normally is not found in water at the concentration required to produce the measured concentrations of radon; that is, in excess of several thousand picocuries per liter.

Although thorium isotopes are not generally significant dissolved radioactive constituents of natural waters, they are formed in the dissolved state by the radioactive decay of the dissolved uranium isotopes U-238 and U-234. Whereas the uranium tends to remain in solution as a stable carbonate complex, the thorium isotopes plate out or are sorbed on rock surfaces. Concentrations of Th-234, U-234 (decay product of Th-234), and Th-230 are much higher on the surfaces of rock fractures that have been exposed to percolating waters than in the bulk of the country rock (J. Rosholt, oral commun., 1977). Radium-226 resulting from the radioactive decay of the sorbed Th-230 probably is the source of much of the dissolved radon found in the thermal waters described in this study.

Wollenberg (1975) and O'Connell and Kaufmann (1976) suggest that radium-226, coprecipitated with calcium, can be a substantial secondary source of radon. At least part of the radon at Alhambra probably is a decay product of radium-226 in calcareous deposits at depth.

No consistent relationship between the concentration of radon and the chemistry or temperature of the water is apparent; however, the concentrations of dissolved radon in samples from flowing wells generally were higher than in samples from the springs (table 1, fig. 2). Spasmodic eruptions of gas from flowing hot well 6, effervescent discharge from flowing warm well 10, and continuous gas bubbles in more active hot springs denote super-saturation of the waters with gas. A 60 mL syringe of gas from hot spring 9 contained the equivalent of 80,000 pCi of radon-222 per liter of gas and a similar sample from flowing hot well 6 contained 130,000 pCi/L. The activity of dissolved radon in corresponding water samples was 7,000 and 11,000 pCi/L, respectively. Radon in the spring waters probably had more time to approach equilibrium with the atmosphere and to dissipate into the surrounding rock and soil at shallow depth than did radon in the more confined well waters.

Major components of the gas from the wells and springs are nitrogen (85-88 percent), oxygen and argon (4-6 percent), and carbon dioxide (8-9 percent). Whereas radon is a noble, nonreactive gas, reactions between carbon dioxide in solution and the rocks probably determine to a large extent the chemical properties of the thermal waters. Although the solubility of carbon dioxide in water varies inversely with temperature, it is retained in solution under hydrostatic head at depth where it can lower the pH of the water and react with the minerals in the rock to form soluble bicarbonates. Conversely, when carbon dioxide gas is liberated from ascending thermal waters as a result of the decline in hydrostatic pressure, the less soluble compounds, particularly calcium carbonate, can precipitate from a saturated solution to seal the conduits or to form travertine deposits at the surface. Precipitation of calcium would leave the waters relatively enriched with respect to the more soluble sodium bicarbonate.

Sodium (in equivalent weights) is more abundant than calcium in the silica-rich rocks associated locally with the hot springs and the radioactive anomalies. The high sodium and bicarbonate content of the thermal waters at Alhambra may result from the reaction:

$$CO_2 + \text{Na silicate} + H_2O = HCO_3^- + Na^+ + \text{H silicate}$$

which represents dissolution of sodium feldspar with the simultaneous conversion of carbon dioxide (CO_2) to bicarbonate (HCO_3^-), release of sodium ion ($Na+$), and formation of clay. Calcium, potassium, silica (SiO_2), and soluble radioactive elements disseminated in the rock would be liberated concurrently. Sodium bicarbonate tends to inhibit precipitation of dissolved uranium (Barker and Robinson, 1963), thereby increasing its mobility.

Subsurface temperatures estimated from the concentrations of sodium, potassium, and calcium in the waters (Fournier and Truesdell, 1974) are about 157°C for hot spring 9 and 161°C for flowing warm well 10. The estimates are based on the assumptions that the ascending water had not interacted with the country rock after it left the geothermal reservoir, that it had not mixed with cooler shallow ground water, nor had any of the three ions precipitated enroute to the point of discharge (Fournier and others, 1974). The difference of about 100°C between the estimated subsurface temperatures and measured surface temperatures suggests mixing. Precipitation of calcium, as almost certainly occurs at Alhambra, would lead to erroneously high temperature estimates, although the error probably is negligible.

The solubility of silica increases with temperature, and the solubility of amorphous silica is much greater than the solubility of primary quartz at the same temperatures. If the dissolved silica in the thermal waters is at equilibrium with primary quartz in the fractured crystalline rock aquifers, subsurface temperatures estimated from the concentrations of silica are about 111°C for hot spring 9 and 115°C for flowing warm well 10 (Fournier and Rowe, 1966). The estimated temperatures would be only about 82° and 87°C, respectively, if the equilibrium were with chalcedony (Fournier and Truesdell, 1974). If the water is cooled by mixing with shallow ground water, temperatures estimated using the cation and quartz geothermometers are nearly equal.

From his studies in Iceland, Arnorsson (1975) concluded that the silica concentration of hot springs associated with low-temperature reservoirs is controlled by the solubility of chalcedony if the reservoir temperature is less than 110°C, by quartz or chalcedony in the range 110° to 180°C, and by quartz at temperatures greater than 180°C. The combined evidence indicates that the reservoir temperature for Alhambra Hot Springs is in the lowest range.

In several abandoned mines near Alhambra, secondary uranium minerals occur along fractures in veins of microcrystalline quartz or as disseminations in the monzonite wall rock "where they have clearly been deposited from circulating ground water" (Thurlow and Reyner, 1952, p. 18). Wright, Bieler, Shulhof, and Emerson (1954) state that the most recent of several varieties of chalcedony found in veinlets and enclosing breccia fragments are the most highly radioactive. However, they confirm the association of primary and secondary uranium minerals with chalcedonic gangue, which led Roberts and Gude (1953) to conclude that little or no migration of uranium has occurred since the primary uranium and chalcedony were deposited by ascending epithermal solutions. Current circulation of thermal water probably is dissolving the radioactive vein material and is unassociated with the primary emplacement and wallrock alteration that apparently took place before late Tertiary time.

The rough correlation between the concentration of silica and radioactivity (fig. 2), the absence of excessive radioactivity in hot springs from similar crystalline rock, and the local concentration of radioactive minerals suggest that leaching of radioactive material from fractured siliceous veins is the principal source of radioactivity in the thermal waters at Alhambra. If the radioactive elements were derived from dissolution of chalcedonic vein deposits intercepted by the Alhambra fault or associated fractures, at least part of the dissolved silica would be derived from the same source. The silica geothermometers then would confirm the association by Mariner, Presser, and Evans (1976) of Alhambra Hot Springs with reservoir temperatures less than 110°C.

CONCLUSIONS

1. High levels of radioactivity occur at Alhambra Hot Springs in the discharges from springs and flowing wells, in calcareous incrustations around the springs, and in silicified zones and chalcedony veins in associated crystalline intrusive rock. High levels of radioactivity are not characteristic of hot springs that issue from fractured crystalline rock in Montana.

2. The thorium and uranium concentrations of the type of acid igneous rocks exposed near the hot springs are higher than the average for the Boulder batholith. The predominance of thorium suggests that a toxic beta-emitting daughter, radium-228, is present in the thermal waters.

3. Gross alpha and beta activities, and the concentrations of radium-228 in thermal waters from springs and flowing wells exceed recommended maximum permissible levels of the U.S. Environmental Protection Agency (1976) for drinking water. High concentrations of radon are dissolved in the water and mixed with other gases at several sites.

4. In general, gross alpha and gross beta activities determined by the USGS exceeded the values reported for corresponding samples by the MDHES. Both laboratories verified anomalously high levels of radioactivity at the same sites and the results were normally proportional. The discrepancies may be attributable to differences in calibration-isotope solutions and analytical procedures.

5. Fractures associated with the Alhambra fault appear to act as conduits for ascending thermal waters that were heated by deep circulation along a thermal gradient of about 0.64°C/100 feet (21°C/km).

6. The chemical composition of the water ranges from calcium bicarbonate type surface water and shallow ground water to more highly radioactive sodium bicarbonate type thermal water with increasing temperature, concentrations of dissolved solids and silica, and sodium-adsorption ratio.

7. Under atmospheric conditions some of the thermal waters are supersaturated with respect to calcium carbonate and carbon dioxide gas. Reactions of dissolved carbon dioxide with rocks in the subsurface probably have released the sodium and calcium ions and produced bicarbonate ion in the water.

8. Loss of carbon dioxide by ascending thermal waters caused deposition of radioactive travertine at the surface, and probably in the subsurface.

9. Radium-226 is the source of radon-222 that is dissolved in the thermal water and discharged with other gases from several springs and wells. The radium-226 probably formed as a decay product of thorium-230 sorbed on fracture surfaces and coprecipitated with calcium carbonate.

10. Dissolution of radioactive chalcedony at depth probably accounts for most of the radioactivity and part of the dissolved silica in the thermal waters.

11. The reservoir temperature probably is less than 110°C, but could be as high as 160°C.

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