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ABSTRACT

Concentrations of selected isotopes in the uranium decay series were determined for samples collected from the Wairakei, Broadlands/Ohaaki and Waiotapu areas. ^{226}Ra concentrations were found to be low (0.05-0.22 dpm/l), similar to values reported in neutral hot springs at Tatun geothermal area, Taiwan, but lower than other geothermal systems (Yellowstone, USA, and Latera, Central Italy) (up to 25 dpm/l). The potential of ^{226}Ra/^{228}Ra ratios for indicating water residence times could not be explored because ^{228}Ra data was not available. ^{222}Rn concentrations are higher and related to steam fractions and CO_2 concentrations. The short half-life (3.8 days) makes ^{222}Rn suitable for estimating residence times of radon in steam, and therefore the distance of travel of steam from its source (e.g., wells WK9 and 52). ^{210}Pb and ^{210}Po concentrations were very low and less than detection limits in many of the Wairakei waters; no residence time applications are apparent for these isotopes.

KEYWORDS

Geothermal systems; Wairakei; Radon; Radium-226; Ohaaki; Radon-222; Polonium-210.

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INTRODUCTION

The Wairakei geothermal system has been studied extensively (e.g. Grindley, 1965). Much has been learned about the natural state of the system (pre-1960) and its subsequent response to exploitation (Allis, 1979; Henley & Stewart, 1983). But problems remain, particularly with understanding processes in the deeper parts of the system and the connections between exploited and non-exploited parts.

In the present work, uranium and thorium decay series isotopes (^{226}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po) have been measured to investigate their use as indicators of fluid residence times in the hot zone of the system, and to find out if ^{210}Po is mobile in the Wairakei system. ^{222}Rn is particularly interesting because it is a gas and potentially useful for investigating the extent of 2-phase conditions in a geothermal reservoir (Horne and Kruger, 1979).

BACKGROUND

Ratios of the uranium and thorium decay-series isotopes and particularly their deviations from equilibrium values have been used as indicators of age in a variety of geological materials (e.g. uranium-thorium ages of shells and corals.). Co-existing isotopes in the decay-series attain equilibrium concentrations if the enclosing medium is left undisturbed for sufficient time. Then, if equilibrium is disturbed by a geochemical process (e.g. preferential solution or deposition), the gradual subsequent approach to re-equilibrium may constitute a "clock" measuring time since the event with time scale dependent on the relative rates of radioactive decay of the isotopes concerned.

Fluid residence times in geothermal systems may be inferred by comparison of radioisotope ratios and radionuclide/stable element ratios in thermal waters to the same ratios in the source material (presumably host rocks). For example, if the ^{210}Pb/Pb ratio in the water is higher than that in the rock, then the excess ^{210}Pb ($t_{1/2}$ = 22.3 years) is inferred to have been derived from decay of the ^{222}Rn ($t_{1/2}$ = 3.8 days) in the water (see decay schemes in Fig. 1). A residence time can therefore be determined based on the ^{210}Pb/Pb ratios, the ^{222}Rn activity and the water/rock ratio. Another example for shorter residence times, would be ^{228}Ra ($t_{1/2}$= 5.75

Fig. 1 **Simplified Uranium-Radium and Thorium decay schemes, omitting some short-lived isotopes.**

(The radioactive half-lives are given below each isotope.)

$^{238}\mathrm{U}^{92}$	→	$^{234}\mathrm{U}^{92}$	→	$^{230}\mathrm{Th}^{90}$	→	$^{226}\mathrm{Ra}^{88}$	→	$^{222}\mathrm{Rn}^{86}$	→	$^{210}\mathrm{Pb}^{82}$	→	$^{210}\mathrm{Bi}^{83}$	→	$^{210}\mathrm{Po}^{84}$	→	$^{206}\mathrm{Pb}^{82}$
4.5x10^{9}yr	longest 24 days	2.5x10^{5}yr		8x10^{4}yr		1620yr		3.8 days	longest 27 min	22.3yr		5 day		138 day		

$^{232}\mathrm{Th}^{90}$	→	$^{228}\mathrm{Ra}^{88}$	→	$^{228}\mathrm{Th}^{90}$	→	$^{224}\mathrm{Ra}^{88}$	→	$^{208}\mathrm{Pb}^{82}$
1.4x10^{10}yr		5.75yr	longest 6 hours	1.9yr		3.6 day	longest 10 hours	

years) normalised to ^{226}Ra ($t_{½}$ = 1620 years) (Clark and Turekian, 1990).

Previous measurement of ^{210}Po concentration have revealed that this element is mobile in some sulphide-rich environments (Harada *et al.*, 1989).

^{222}Rn is produced by α-decay of ^{226}Ra. In a closed system, ^{222}Rn would be in radioactive equilibrium with its parent, but measurements in geothermal areas have shown that ^{222}Rn is greatly in excess of equilibrium in geothermal fluids (Whitehead, 1979). Clearly there is preferential loss of radon from the rock and Battaglio *et al.* (1992) show that this occurs mainly by recoil. Movement of fluid out of the pores of the rock allows the radon to migrate. The radon concentrates in the steam phase if steam is present; the equilibrium separation factor ($A = C_s/C_w$, where C_s and C_w are the concentrations of ^{222}Rn in steam and water respectively in nCi/kg) being 550 at 220°C and 1480 at 180°C (Potter and Clynne, 1978).

SAMPLING & MEASUREMENT

Samples were collected from geothermal wells at the Wairakei geothermal area (Fig. 2), from two wells at the Broadlands/Ohaaki geothermal area and from Champagne Pool (a large natural hot pool) at the Waiotapu geothermal area. The main focus was on Wairakei, but samples from other areas were collected for comparison.

Well samples were collected in a variety of ways depending on the wellhead equipment (see sample types in Table 1). Where possible, water samples were collected under pressure from the water phase outlet (waterline) and steam from the steam phase outlet (steamline) of wellhead separators. These were cooled and condensed respectively in a cooling coil. In other cases, water phase samples were collected at atmospheric pressure from weirboxes after they had cooled to 100°C by loss of steam (flashing). When there were no wellhead separators, the 2-phase mixture within the wellpipe was discharged to the atmosphere and 100°C water collected (flashed total discharge). The groundwater well WK228/0 was sampled using a downhole sampler and water from Champagne Pool was collected in a dipper.

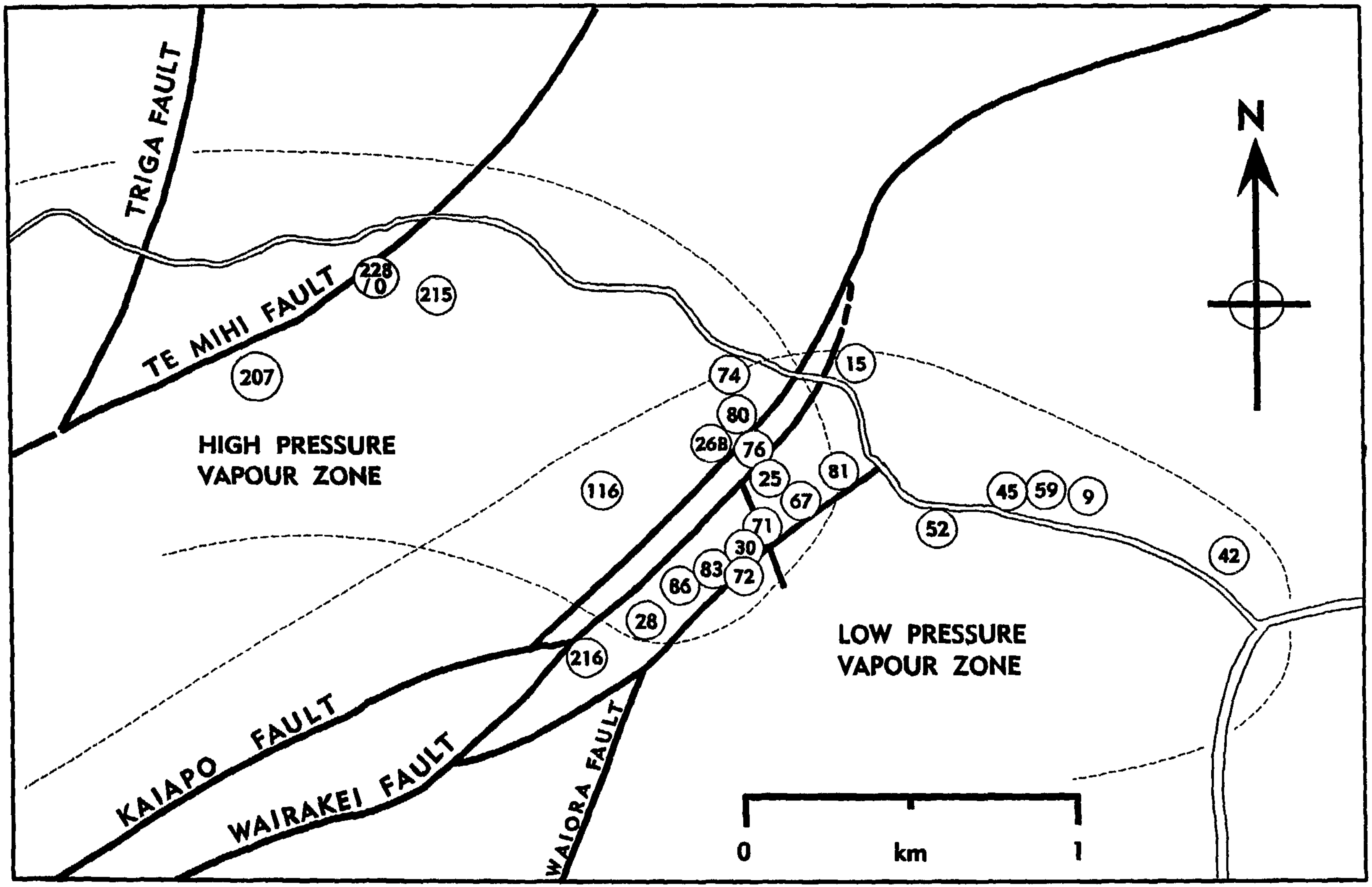

Fig. 2 : Map of the production area of Wairakei showing well locations and major faults.

TABLE 1: Concentrations of some natural decay-series isotopes in Wairakei waters

Well	Sample Type +	^{226}Ra dpm/l	^{222}Rn dpm/l*	^{210}Pb dpm/l	^{210}Po dpm/l
WK9	wbx	0.15	13926	<0.08	0.38
WK26B	csg	0.1	2937	<0.23	0.14
WK28	csg	0.17	2999	0.18	0.08
WK52	wbx	0.16	8308	<0.06	0.15
WK59	wbx	0.13	7370	<1.00	0.19
WK67	cwl	0.06	373	<0.11	0.12
WK76	csg	0.05	2691	0.16	0.14
WK80	wbx	0.17	25774	<0.04	0.09
WK80	csl	-	-	1.35	1.53
WK86	csg	0.22	16903	0.25	0.06
WK36	csl	-	-	0.71	0.34
WK116	wbx	0.14	1210	0.4	0.1
WK207	ftd	0.19	42	<0.22	0.12
WK215	ftd	0.22	11575	<0.18	0.06
WK228/0	dh	0.24	0	<0.06	0.16
BR22	ftd	0.18	251	0.2	12.8
BR25	ftd	0.21	150	<0.16	0.9
Champagne Pool	dipper	0.3	355	<0.05	0.73

+wbx - weir box water
csg - cooled water phase sample from sight glass
cwl - cooled water phase sample from water line
csl - condensed steam phase sample from steam line
ftd - water from flashed total discharge
dh - samples collected downwell with the downhole sampler

* ^{222}Rn concentrations were for condensed steam as described in the text.

The water samples were analysed for radium-226, lead-210 and polonium-210, and two steam samples for lead-210 and polonium-210. Radioisotope measurements of polonium-210 and lead-210 were carried out by electroplating and α or ß counting. Radium was coprecipitated with barium sulphate and determined via PERALS spectrometry (Burnett and Tai, 1992). The results are expressed as concentrations in the downhole water.

Samples for radon-222 were collected by condensing steam in a cold trap and allowing the gas to bubble through it. Only a fraction of the radon was retained in the condensed steam, but this gives a qualitative measure of the total radon-222 content. Comparison with earlier radon measurements (Whitehead, 1979) indicates that about 10% of the radon was retained in the condensed steam.

RESULTS AND DISCUSSION

Results are given in Tables 1 and 2. Table 1 gives the decay-series isotope results expressed as concentrations in the downhole water for all except the radon which is concentration in condensed steam. Table 2 includes data relevant to the Wairakei field obtained at the same time as the decay-series samples. The steam fractions of fluid feeding the wells were determined from the discharge enthalpies and downhole temperatures. (The latter were estimated from the silica contents assuming equilibrium with quartz.) Chloride and stable isotope concentrations of the downhole water are also given. These were used to determine the proportions of groundwater in the water fraction of the discharge after allowing for steam loss processes underground (see description below). CO_2 concentrations are given as weight percent of the total discharge. CO_2 (like radon) emerges predominantly in the steam phase.

The Wairakei system has been heavily exploited for electricity production since the early 1950's, causing extensive pressure drawdown and formation of a steam cap (or more precisely, a 2-phase zone with mobile steam) in the originally mainly liquid geothermal aquifer. Monitoring of pressures in the steam cap revealed that there were two main vapour zones (Fig. 2); a low pressure zone covering both the eastern and western exploited areas and extending to the west south west, and a deeper high pressure zone under the low pressure zone in the western exploited zone extending to the west north west (Electricorp, 1990). The

TABLE 2: Physical, chemical and isotopic data for Wairakei and other waters

Well	Enthalpy kJ/kg	Tquartz °C	Steam fraction	Chloride mg/kg	$\delta^{18}O$ ‰	δD ‰	Tritium TR	Dilution	CO_2 Wt.%
WK9	1650	169	0.45	309	-6.97	-43.0	0.14± .07	0.81	0.27
WK26B	945	228	-0.02	1483	-6.10	-45.0	-	0.10	0.003
WK28	995	231	0.00	1448	-5.99	-44.3	0.12 .06	0.08	0.02
WK52	1720	173	0.48	431	-7.01	-43.5	0.26 .08	0.73	0.21
WK59	950	206	0.03	1348	-6.20	-43.8	0.14 .06	0.16	0.013
WK67	975	236	-0.02	1416	-5.97	-43.9	0.16 .07	0.12	0.004
WK76	940	230	-0.02	1363	-6.16	-44.5	0.16 .07	0.12	0.006
WK80	1500	183	0.36	1235	-5.64	-42.5	0.08 .06	0.28	0.11
WK86	1105	226	0.07	1465	-5.86	-44.7	0.03 .06	0.11	0.056
WK116	1020	234	0.00	1620	-5.97	-45.1	-	0.02	0.005
WK207	1070	232	0.04	1705	-5.89	-46.0	0.07 .04	-0.01	0.015
WK215	1130	228	0.08	1727	-5.88	-45.5	0.10 .07	-0.02	0.014
WK228/0	-	-	0.00	21	-7.35	-46.2	1.60(e)	1	0
BR22	1220	268		1232	-4.42	-41.0	0.03 .05	0.00	0.59
BR25	1577	269		960	-4.90	-43.0	-	0.25	2.79
Champagne Pool	75°C	218	-	1898	+3.06	-22.4	-	-	-

$\delta^{18}O$ ‰ = $[(^{18}O/^{16}O)_{sample}/(^{18}O/^{16}O)_{V\text{-}SMOW} - 1] \times 1000$. Similarly for δD.

decrease in pressure in the aquifer has allowed penetration of groundwater down through the vapour zones into the liquid region. The estimated fractions of groundwater dilution of the water phase are given in Table 2 for the wells sampled.

Wells WK9 and 52 draw steam and water from the low pressure vapour zone at about 150 m depth (see steam fractions in Table 2) and discharge dilute waters (containing about 80% heated groundwater). WK80 draws steam from the high pressure vapour zone and contains 30% groundwater. Most of the wells in the western area draw from deeper levels and have a groundwater dilution of about 10% with small proportions of steam from the high pressure vapour zone, while WK59 in the heavily degraded eastern area has 16% groundwater. WK116, 207 and 215 in the less-exploited western area have zero groundwater dilution.

^{226}Ra concentrations in the water are very low (0.05-0.22 dpm/l). In comparison, concentrations in hot spring waters at Yellowstone (Clark & Turekian, 1990) ranged from 0.1 to 24.8 dpm/l, while those in waters from geothermal wells at Latera, Central Italy (Battaglio *et al.*, 1992) had 5-10 dpm/l; these higher concentrations probably indicate that the source rocks are more radioactive. Concentrations in hot springs at Tatun, Taiwan (0.01-0.30 dpm/l; Tai, 1992) were similar to those at Wairakei, except for two acid springs (pH ~1) which had 1.3 and 2.5 dpm/l; probably because acid conditions promote greater rock dissolution and less adsorption of radium on rock surfaces. The ^{228}Ra/^{226}Ra ratio was used by Clark and Turekian (1990) to estimate a fluid residence time of 500 years in the hot zone of the Norris-Mammoth Corridor, Yellowstone National Park, but could not be investigated here because ^{228}Ra was not measured.

^{222}Rn concentrations are considerably higher than those of other radioelements at Wairakei. The current data along with earlier radon measurements from Whitehead (1979) and Horne and Kruger (1979) are listed in Table 3 as nCi/l (= dpm/l × 0.45/1000). The current data have been multiplied by a factor of 10 to correct for loss of radon during sample collection. The ^{222}Rn concentrations are plotted against aquifer steam and CO_2 contents in Figs 3 and 4. A general trend of increasing radon with increasing steam and CO_2 can be seen, reflecting the fact that radon preferentially occupies the vapour phase. In fact, two trends have been identified; an upper trend involving wells associated with the deeper (high pressure) vapour

TABLE 3: Wairakei radon data

Well	Date	Radon-222 nCi/l cond	Steam f Sep	Steam f Aq	CO2 Wt %	Enthalpy kJ/kg	Source
15	01-Oct-76	100.9	0.71	0.69	0.32	2154	a
15	25-Feb-76	188.7	0.71	0.69	0.32	2154	a
28	26-Feb-76	0.12	0.15	0.01	0.008	1051	a
42	28-Jul-78	45.5	0.38	0.29	0.068	1447	a
42	27-Feb-76	49.1	0.34	0.25	0.068	1368	a
42	27-Feb-76	45.5	0.34	0.25	0.068	1368	a
45	27-Jul-78	83.2	1.00	1.00	0.32	2779	a
45	27-Jul-78	111.4	1.00	1.00	0.32	2779	a
74	25-Feb-76	11.1	0.14	0.02	0.009	1042	a
81	27-Jul-78	0.2	0.11	-0.03	0.0012	991	a
216	27-Jul-78	54.1	1.00	1.00	0.24	2784	a
30	04-May-79	0.85	0.10	-0.03	0.0017	982	b
46	04-May-79	4.2	0.17	0.02	0.007	1026	b
71	04-May-79	0.95	0.10	-0.04	0.002	956	b
72	04-May-79	6.9	0.37	0.28	0.073	1519	b
80	07-Mar-79	139	0.36	0.32	0.137	1426	b
83	04-May-79	2.5	0.11	-0.02	0.012	1009	b
86	07-Mar-79	3.4	0.26	0.12	0.059	1209	b
9	01-May-90	63	0.51	0.45	0.27	1650	c
26B	02-May-90	13	0.12	-0.02	0.003	945	c
28	02-May-90	14	0.13	0.00	0.02	995	c
52	01-May-90	37	0.54	0.48	0.21	1720	c
59	01-May-90	33	0.17	0.03	0.013	950	c
67	01-May-90	1.7	0.13	-0.02	0.004	975	c
76	02-May-90	12	0.12	-0.02	0.006	940	c
80	02-May-90	116	0.39	0.36	0.11	1500	c
86	03-May-90	76	0.20	0.07	0.056	1105	c
116	02-May-90	5.5	0.16	0.00	0.005	1020	c
207	02-May-90	0.2	0.16	0.04	0.015	1070	c
215	02-May-90	52	0.20	0.08	0.014	1130	c
228/0	03-May-90	0	0.00	0.00	0	55	c

Sources of data:		
	a	Whitehead (1979)
	b	Horne & Kruger (1979)
	c	This work

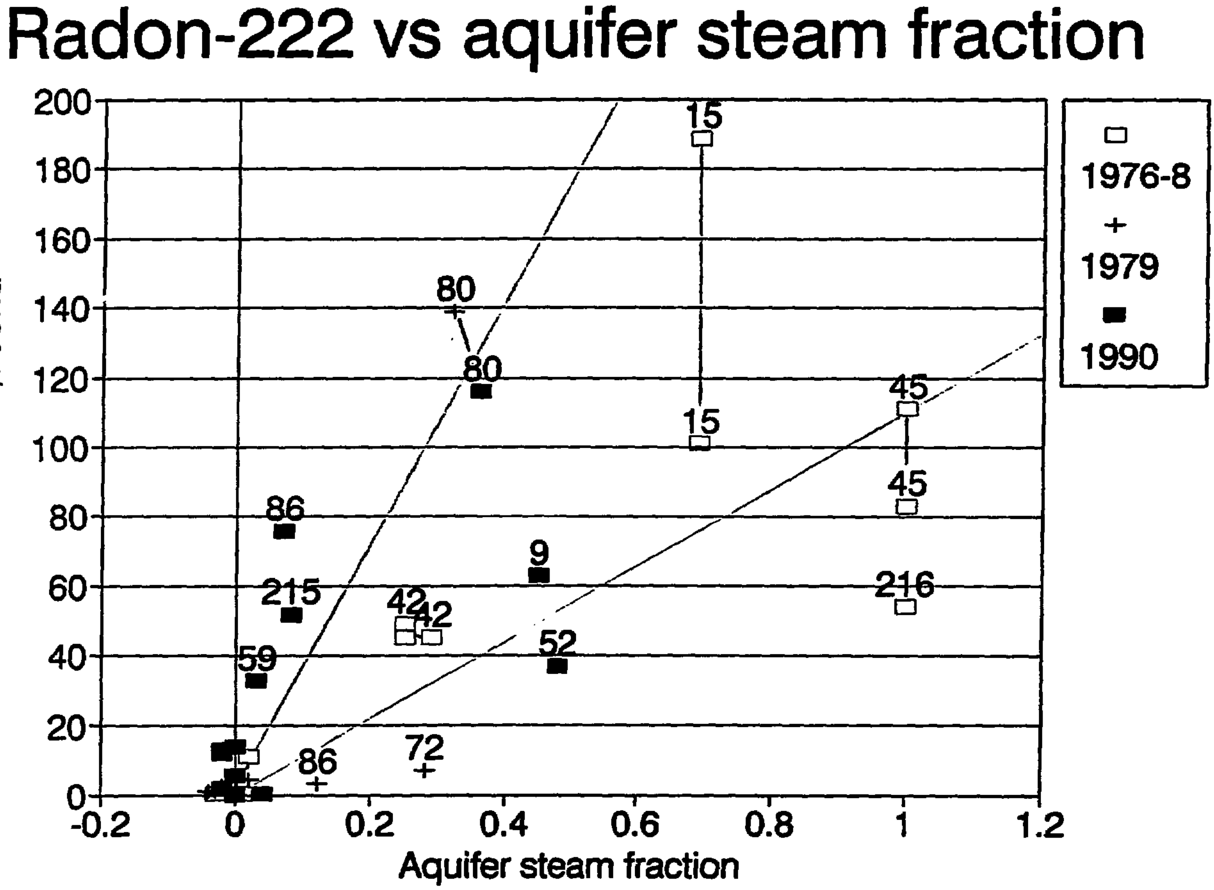
Radon-222 vs aquifer steam fraction
Radon-222 nCi/l cond
200
180
160
140
120
100
80
60
40
20
0
-0.2
0
0.2
0.4
0.6
0.8
1
1.2
Aquifer steam fraction
1976-8
1979
1990
15
15
80
80
45
45
86
9
216
215
42
42
59
52
86
72

Fig. 3

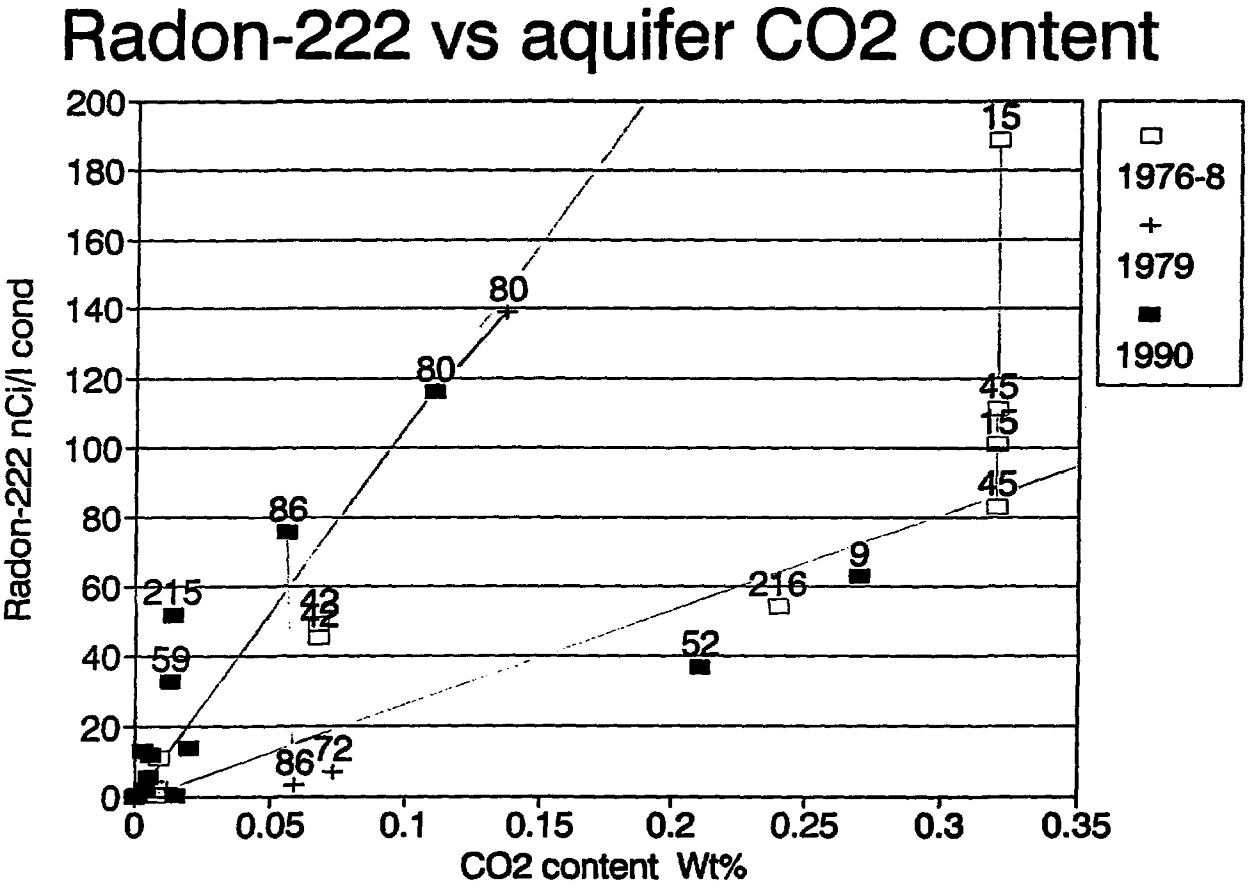
Radon-222 vs aquifer CO2 content
Radon-222 nCi/l cond
200
180
160
140
120
100
80
60
40
20
0
0
0.05
0.1
0.15
0.2
0.25
0.3
0.35
CO2 content Wt%
□ 1976-8
+ 1979
■ 1990
15
80
80
45
15
45
86
9
216
215
43
42
52
59
72
86

Fig. 4

zone (wells WK59, 80, 86, 215) and a lower trend associated with the low pressure vapour zone (wells WK9, 45, 52, 216). Wells 15 and 42 lie between the trends. Although both zones are supplied by steam from the deep liquid system, the low pressure zone has lower radon content implying longer residence times and therefore greater radioactive decay of the radon, especially on the eastern side of the field (e.g., WK9, 52, 45). Residence times of a few days longer than in the high pressure zone are indicated. CO_2 concentrations correlate quite well with the downhole steam content (see Fig. 5), except for several wells drawing steam from the low pressure zone (WK9, 15, 52). The relatively high CO_2 concentrations in these wells are attributed to condensation of steam because of groundwater input (e.g., water phase discharged from wells WK9 and 52 contain 80% steam-heated groundwater).

In comparison with Broadlands, Wairakei wells show a very large range in radon contents. At Wairakei the range is 0.2-189 nCi/l (Table 3), while at Broadlands it is 1.3-10.7 nCi/l (Whitehead, 1979). The present samples gave 1.1 nCi/l for BR22 (previously 2.6-10.7 nCi/l) and 0.7 nCi/l for BR25 (previously 1.3 nCi/l). The source of the high values at Wairakei is an interesting question. It is certain that radon emanating from the rock (because of recoil) is collected initially into water. This is because by far the largest proportion of the aquifer rock is in intimate contact with water phase, even within the steam zone, since water occupies the smallest crevices because of capillary forces. Steam, because of its lower density, occupies the larger voids when it is present above a threshold proportion. The extensive drawdown at Wairakei has led to continual drainage of water, initially from the larger voids but increasingly from finer and finer crevices. It is probable that water flowing from intimate contact with the rock in these crevices carries high radon content, which then passes preferentially into steam when flashing occurs. Broadlands has not been affected by the same degree of pressure drawdown.

Both ^{210}Pb and ^{210}Po are very low at Wairakei, indicating that these isotopes are removed from solution by deposition or exchange mechanisms. Interestingly, both isotopes are higher in the steam samples (i.e. the csl samples from wells WK80 and 86; see Table 2), probably as a result of ^{222}Rn decay.

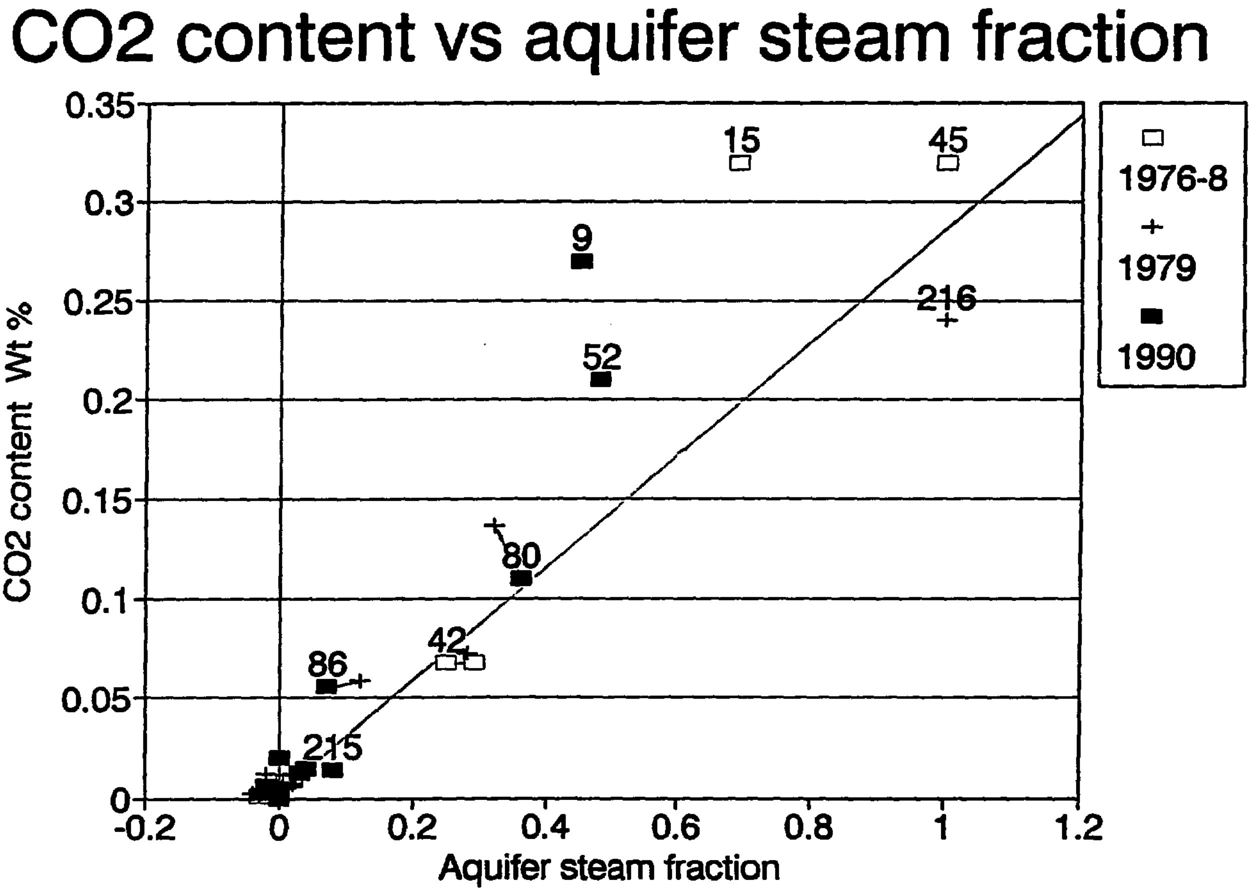
CO2 content vs aquifer steam fraction
CO2 content Wt %
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
-0.2
0
0.2
0.4
0.6
0.8
1
1.2
Aquifer steam fraction
15
45
9
216
52
80
42
86
215
1976-8
1979
1990

Fig. 5

Two samples were collected from the Broadlands/Ohaaki geothermal area. Geothermal waters from wells west of the Waikato River (BR22) have relatively high chloride concentrations and low fractions of steam. Wells east of the river (BR25) have lower chloride concentrations and therefore apparently greater groundwater dilutions, but higher steam fractions. The reasons for these differences are not entirely clear. The decay-series isotope concentrations are interesting. ^{226}Ra contents are very low, like those at Wairakei, indicating rapid deposition in minerals (calcite). The ^{222}Rn is low and uniform, in agreement with earlier measurements (Whitehead, 1979). This indicates that radon is not being released by the rock. Radon is low even for the high steam well (BR25) in contrast to high steam wells at Wairakei. ^{210}Pb is very low at Broadlands, but ^{210}Po shows one quite high value. BR22 chemistry is affected by an organic chemical, which is injected downwell to retard calcite deposition within the well, but this is unlikely to have affected the polonium. The ^{226}Ra shows no effect of it. It is more likely that polonium is mobile at Broadlands because of the relatively higher H_2S environment. More measurements would be useful at Broadlands to confirm or disprove these suggestions.

Champagne Pool is a large flowing spring (diameter ~30m) at Waiotapu geothermal area. The pool waters overflow a sinter rim, which has been found to contain significant quantities of antimony and arsenic sulphides (also up to 80 ppm gold and 175 ppm silver). The water has the highest chloride concentration of any spring in the area and is sourced from deep water, probably without groundwater dilution. The stable isotope composition is quite enriched in heavy isotopes showing that considerable evaporation has occurred at or on the way to the surface. The spring has a natural ebullience of gases. ^{226}Ra concentration is slightly higher than in the other samples, while ^{222}Rn is low as expected from the visible evidence of loss of gas. Both ^{210}Pb and ^{210}Po are low, but polonium is higher than in the Wairakei samples.

SUMMARY

Concentrations of some isotopes in the uranium decay series were determined for samples collected from the Wairakei, Broadlands/Ohaaki and Waiotapu geothermal areas and the results discussed in terms of residence time applications. Use of ^{228}Ra/^{226}Ra and radon isotopes are considered the most promising techniques for further work.

^{226}Ra concentrations were found to be very low at all areas but similar to values measured at Tatun geothermal area, Taiwan. Other geothermal systems (Yellowstone, USA, and Central Italy) had higher concentrations. ^{228}Ra/^{226}Ra ratios have been used as indicators of fluid residence times elsewhere, but ^{228}Ra values are not available for Wairakei.

^{222}Rn concentrations are higher and related to steam fractions and CO_2 concentrations. Two trend lines were identified and related to the low and high pressure vapour zones. The short half-life (3.8 days) makes ^{222}Rn suitable for estimating residence times of radon in steam, and therefore the distance of travel of steam from its source (e.g. wells WK9 and 52). This application needs to be refined with further work.

^{210}Pb concentrations were less than detection limits in many of the Wairakei waters, although the two steam samples had considerably higher concentrations. It appears that Pb is rapidly removed from the water by mineral deposition or the natural tendency of ^{210}Pb to plate out, and thus ^{210}Pb cannot be used for determining residence times because of its low concentration. ^{210}Po concentrations were also low in water and higher in steam, except in one well at Broadlands/Ohaaki (BR22) in which the water had a much higher value. This is thought to be because the fluid had a high H_2S concentration.

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