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Lead-210 in Southern California Groundwaters

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ABSTRACT

As part of a geochemical monitoring program for earthquake prediction studies in Southern California, both radon and helium in groundwaters were measured monthly at the network sites from 1974 to 1985. Along with this monitoring program, lead-210 and radium-226 were also measured at most of the network sites, including those in the Palmdale area, for their spatial variation and correlation with radon during the first few years. These measurements show that both the ^{210}Pb and ^{226}Ra activities at the same site are comparable, but they are only about 10^{-4} times the radon activities. The extremely high activities of radon relative to those of ^{226}Ra suggest that radon diffuses into the circulating groundwaters from the ambient rocks. The low activities of ^{210}Pb relative to those of radon imply that either ^{210}Pb produced by radon decay in the groundwaters is removed rapidly by adsorption onto fractured rock surfaces or radon is injected into the groundwaters only at shallow depths with a very short residence time.

An apparent model age of the groundwater since the injection of ^{222}Rn can be calculated from the $^{210}Pb/^{222}Rn$ activity ratio assuming no ^{210}Pb present in the groundwater when ^{222}Rn was injected. The calculated model ages, ranging from 3 hours to 9 days, are indeed very short compared to any estimate of groundwater circulation times. If ^{210}Pb is removed from the circulating water by particulate scavenging and/or adsorption onto the fractured rock surfaces in contact with the water, then a typical residence time for ^{210}Pb in the water can also be calculated based on the $^{210}Pb/^{222}Rn$ activity ratio. This calculated residence time for ^{210}Pb is quite comparable to the apparent model age of the groundwater since the injection of radon. However, the extremely low $^{210}Pb/^{222}Rn$ activity ratios are more likely due to rapid removal of ^{210}Pb from the waters by adsorption onto the fractured rock surfaces or particulate matter.

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1. INTRODUCTION

As part of the Earthquake Hazard Reduction Program sponsored by the U.S. Geological Survey, radon , helium and other dissolved gases in groundwaters were monitored as possible fluid-phase precursors to earthquakes in the hot springs and thermal wells along the Elsinore, San Jacinto and San Andreas faults between San Bernardino and the Mexican border in California for the decade from 1974 to 1985. The monitoring work was further extended to include the Palmdale area during 1977 and 1978. The sampling sites together with the major faults are shown in Figure 1.

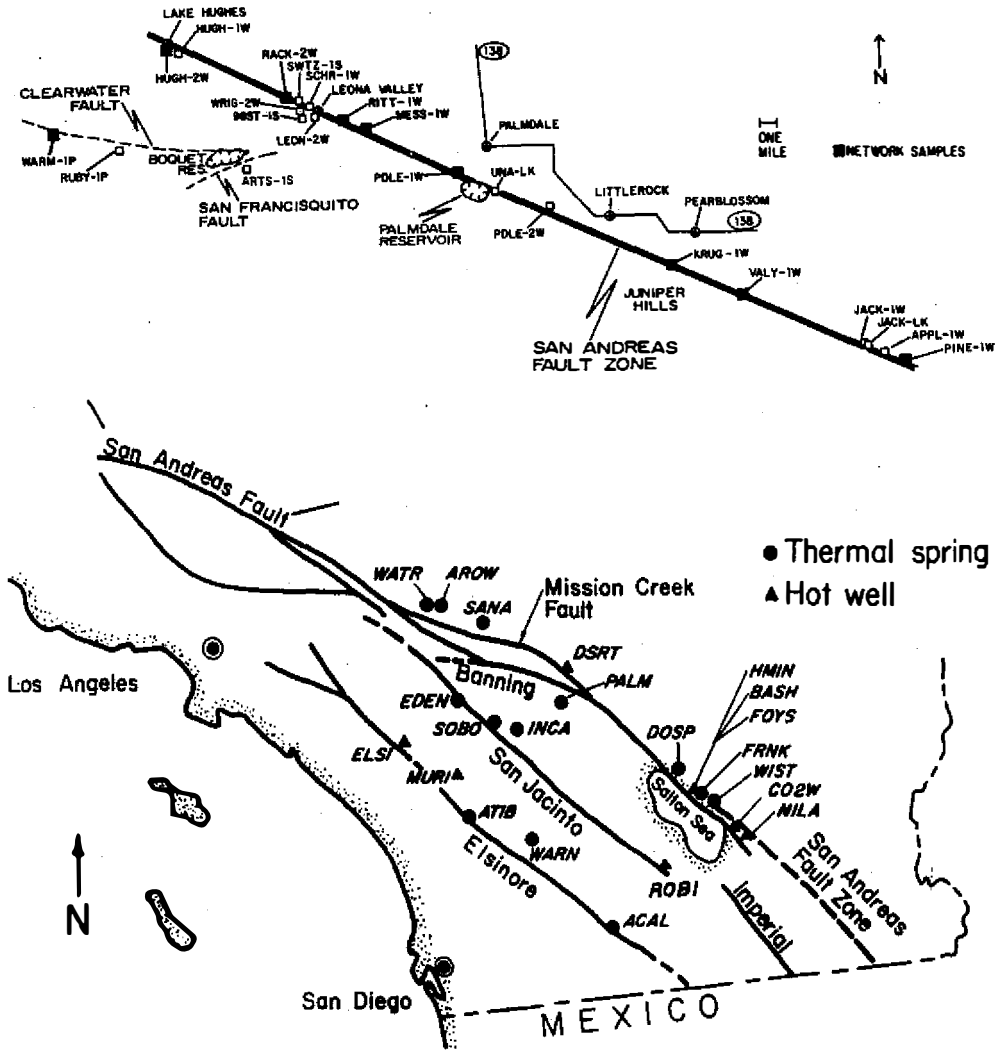

Fig. 1. Radon and helium monitoring sites along the major fault zones in southern California. The Palmdale area north of Los Angeles along the San Andreas is included in an enlarged scale.

Samples of 20-liter size were collected from most of the primary sites between 1975 and 1979 for ^{210}Pb and ^{226}Ra measurements so that the spatial variations of these nuclides as well as their correlations with radon might be examined. These measurements were all accompanied by our routine radon and helium monitoring. The measurements indicated that ^{226}Ra activities had large spatial variations and were 2 to 5 orders of magnitude *lower* than the ^{222}Rn activities of the same sites at the same sampling time (Chung, 1981). The activities of ^{210}Pb were comparable to those of ^{226}Ra within two orders of magnitude but were about 10^{-4} times the ^{222}Rn activities at the corresponding sites. These data and their relationships allowed us to characterize the groundwaters in Southern California fault systems. The spatial variations of ^{226}Ra and ^{222}Rn and their relationships to temperature and conductivity have been discussed (Chung, 1981). This paper presents the ^{210}Pb results together with the associated ^{222}Rn activities and their ratios applied for estimating apparent model ages or residence times for ^{210}Pb in the circulating groundwaters.

2. ^{210}Pb MEASUREMENTS AND RESULTS

Groundwater samples for ^{210}Pb measurements were collected in 20-liter glass bottles and immediately purged with air to remove all the radon. The air-stripped samples were then transferred into plastic containers, acidified to pH about 2, and a stable Pb carrier in a solution of $Pb(NO_3)_2$ and $FeCl_3$ was added to each sample for isotopic equilibrium. The sample was processed in the laboratory following the technique described by Craig *et al.* (1973) and Applequist (1974). The precision of measurements was generally about ±5%.

The ^{210}Pb and ^{222}Rn data collected on the same dates are given in Table 1. Most of the ^{226}Ra and ^{222}Rn data presented earlier (Chung, 1981) are also listed in the table for comparison with the ^{210}Pb data. ELSI (Elsinore Hot Spring), MURI (Murrieta Hot Spring) and ATIB (Agua Tibia Spring) are located along the Elsinore fault (see Figure 1). At ELSI and MURI, ^{210}Pb is about ten times higher than ^{226}Ra, but at ATIB they are quite comparable. EDEN (Eden Hot Spring), at the northern end of the San Jacinto fault, shows significant temporal variations in all nuclides: higher ^{210}Pb reflects higher ^{222}Rn, and ^{226}Ra is about 5 orders of magnitude *smaller* than ^{222}Rn and 2 orders *smaller* than ^{210}Pb. Spatial variations of ^{222}Rn and ^{210}Pb appear to be quite independent of ^{226}Ra variations.

Along the Misson Creek fault, AROW (Arrowhead Hot Spring) and DSRT (Desert Hot Spring) show comparable ^{210}Pb level although ^{222}Rn at DSRT is twice as much as at AROW. In the Salton Sea area along the San Andreas fault zone, large spatial variations of all nuclides are observed: HMIN (Hot Mineral Well) shows about 2 orders of magnitude higher than NILA (Niland Slab Well) in ^{210}Pb, ^{222}Rn and ^{226}Ra. These large differences within a small area along the same fault probably reflect different lithology and mineral composition of the fractured rocks in contact with the groundwaters.

^{210}Pb levels in the Palmdale area are fairly constant as shown in Table 1. The sampling sites are listed in the order from the northwest to the southeast along the San Andreas (Figure 1). ^{222}Rn variations among the sites are within a factor of 7. In the Palmdale area ^{226}Ra is in general *higher* than ^{210}Pb, while in the Southern Network ^{226}Ra is *lower* than ^{210}Pb (except for HMIN and NILA where ^{226}Ra is two orders of magnitude higher than ^{210}Pb).

Table 1. ^{210}Pb, ^{226}Ra and associated ^{222}Rn data in Southern California groundwaters.

Site	Date	Activity (dpm/kg) ^{210}Pb	^{222}Rn	^{226}Ra	$(^{210}Pb/^{222}Rn)\times10^4$	"Age" (days)
			Southern Network			
ELSI-1W	10/22/75	0.426	229		18.6	8.82
	12/21/76		401	0.058		
MURI-1W	4/23/76	0.167	308		5.42	4.22
	12/21/76		216	0.015		
ATIB-1W	11/18/75	0.060	148		4.05	3.42
	12/14/76		96	0.069		
EDEN-1P	11/20/75	2.55	8,760		2.91	2.65
	2/16/77		7,320	0.092		
	7/11/78	4.08	10,700	0.065	3.81	3.27
AROW-1P	4/28/76	0.141	321		4.39	3.63
	12/7/79		292	0.083		
DSRT-1W	10/30/75	0.171	635		2.69	2.49
	12/7/79		572	0.056		
HMIN-1W	11/1/75	2.66	41,600*		0.639	0.70
	1/8/76		46,700*	99.9		
NILA-2W	5/21/76	0.095	312*	3.41	3.04	2.74
			Palmdale Area			
WARM-1P	7/12/78	0.051	293	0.303	1.74	1.73
HUGH-2W	7/12/78	0.032	~900†	0.111	0.356	0.40
RACK-2W	7/12/78	0.026	2,090	0.199	0.124	0.14
RITT-1W	7/13/78	0.017	305	0.025	0.557	0.62
MESS-1W	7/12/78	0.028	910	0.060	0.308	0.35
PDLE-1W	7/12/78	0.035	514	0.036	0.681	0.74
KRUG-1W	7/13/78	0.029	498	0.082	0.582	0.64
VALY-1W	7/13/78	0.025	1,660	0.431	0.151	0.17
PINE-1W	7/13/78	0.012	734	0.035	0.163	0.19

* Excess radon, i.e. radium corrected

† Sample not collected, mean value from other dates of collection

3. $^{210}Pb/^{222}Rn$ ACTIVITY RATIO AND MODEL AGE

The $^{210}Pb/^{222}Rn$ activity ratio varies by 2 orders of magnitude from 10^{-3} to 10^{-5} (Table 1). In the Palmdale area, the ratio is smaller and more uniform at about 10^{-5}. In the Southern Network, the ratio is more variable and generally at about 10^{-4}.

In water with an initial ^{222}Rn activity of A^o_{Rn} but with no ^{210}Pb present, the ^{210}Pb to ^{222}Rn activity ratio will reach unity in about 42 days in a closed system, when ^{210}Pb activity reaches its maximun. After 42 days, ^{210}Pb will begin to decay, but at a much slower rate than ^{222}Rn, and so the ^{210}Pb to ^{222}Rn activity ratio will approach infinity very quickly. In this closed system, the activities of ^{222}Rn and ^{210}Pb at any lapse time t are governed by the equations:

$$A_{Rn} = A^{\circ}_{Rn} e^{-\lambda Rn^{t}} \tag{1}$$

$$A_{Pb} = \frac{\lambda_{Pb} A^{\circ}_{Rn}}{\lambda_{Rn} - \lambda_{Pb}} (e^{-\lambda_{Pb} t} - e^{-\lambda_{Rn} t}) \tag{2}$$

where λ_{Rn} and λ_{Pb} denote decay constant for ^{222}Rn and ^{210}Pb, respectively. However, since λ_{Pb} (8.510^{-5} d^{-1}) is orders of magnitude smaller than λ_{Rn}(0.1812 d^{-1}), $\lambda_{Rn} - \lambda_{Pb} \rightleftharpoons \lambda_{Rn}$, and $\lambda_{Pb}t$ is very small for t even on the order of 100 days (0.0085), so that $e^{-\lambda_{Pb} t} \rightleftharpoons 1$. Thus equation (2) can be approximated as :

$$A_{Pb} = \frac{\lambda_{Pb}}{\lambda_{Rn}} A^{\circ}_{Rn} (1 - e^{-\lambda_{Rn} t}) \tag{3}$$

Based on equations (1) and (3) and the observed activity ratio, we can calculate an apparent "model age" of the groundwater and its initial ^{222}Rn activity, A°_{Rn}, when the groundwater was injected with ^{222}Rn and was free of ^{210}Pb. Denoting the ^{210}Pb/^{222}Rn activity ratio as R and rearranging the terms, we have:

$$t = \frac{1}{\lambda_{Rn}} ln(1 + R \cdot \frac{\lambda_{Rn}}{\lambda_{Pb}}) \tag{4}$$

Using equation (4), we have computed the model ages for all the observed ratios. These model ages are listed in Table 1. Except for ELSI which has the longest model age of 8.8 days, all the sites have R values between 10^{-3} to 10^{-5}, corresponding to model ages of 6.3 days to 2.8 hours. Palmdale values tend to cluster in a small range with model ages between 3 hours and 1.7 days. These ages represent the lapse time required for ingrowth of all the observed ^{210}Pb by ^{222}Rn decay assuming no initial ^{210}Pb content and no gain or loss of these nuclides during this time. These ages are very short by any estimate of groundwater circulation times.

The problem in the model lies in the assumptions that ^{222}Rn was injected some time with an "initial" activity, A°_{Rn}, and that nothing happened other than radiodecay in the (closed) system. It is conceivable that ^{222}Rn must have been added into the system continually, and so the ^{210}Pb to ^{222}Rn activity ratio may not provide any age since the "clock" has been reset constantly. Similarly, ^{210}Pb in the circulating water may not have a zero "initial" activity since it must have been subject to a continuous input (by decay of ^{222}Rn) as well as removal from the circulating water probably by adsorption onto fractured rock surfaces and/or particle scavenging. Scavenging of ^{210}Pb by particulate matter is commonly observed in the oceans (*e.g.* Craig *et al.*, 1973; Somayajulu and Craig, 1976; Bacon *et al.*, 1976) and also occurred in groundwaters (*e.g.* Krishnaswami *et al.*, 1982).

Since the groundwater typical residence times are much greater than the apparent model ages, and ^{222}Rn must have derived from the deep as well as the shallow regions, the model age based on the activity ratio may serve to indicate a short residence time for ^{210}Pb (and perhaps reactive elements also) in the circulating groundwaters rather than the residence time of the groundwaters. If ^{210}Pb is removed rapidly from the circulating water by adsorption onto the surfaces of fractured rocks and/or by scavenging of particulates as mentioned, one can calculate the removal rate of ^{210}Pb based on the activity ratio assuming at steady state. The

residence time of ^{210}Pb calculated with respect to such removal processes in the groundwaters is quite similar to the apparent model age within the ^{210}Pb/^{222}Rn activity ratios observed. Short residence time for reactive elements such as thorium and lead in groundwaters was also observed elsewhere (*e.g.* Krishnaswami *et al.*, 1982).

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