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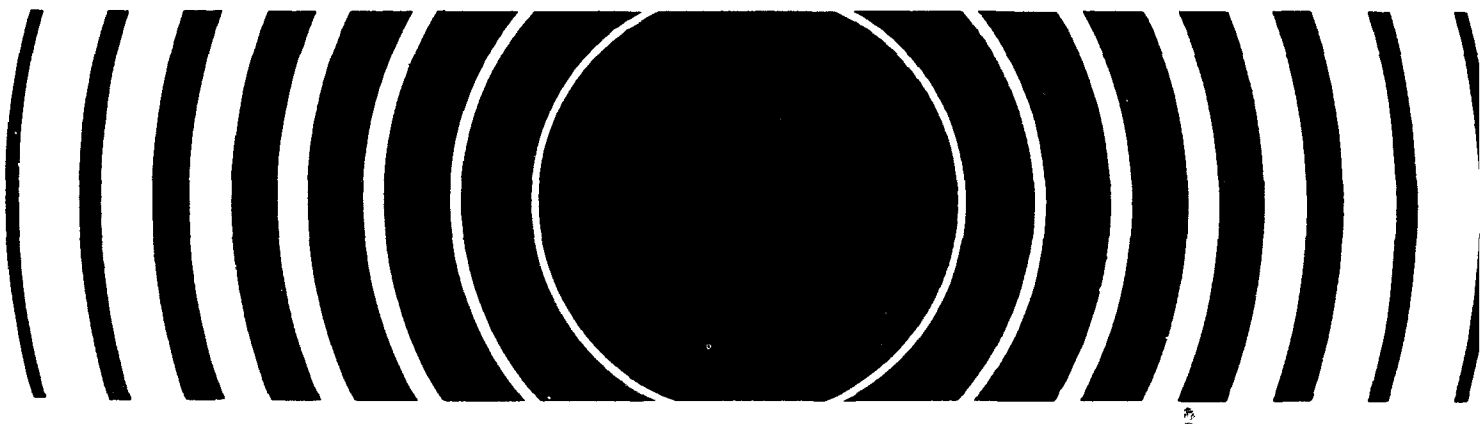

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METHODS AND RESULTS OF EPA'S STUDY OF RADON IN DRINKING WATER

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December 1983

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ACKNOWLEDGEMENTS

Special thanks are extended to all the state health department personnel who collected most of the samples reported in this paper. Without their help this study would have been impossible to conduct. Special appreciation is also extended to Kitty Newman for her patience and steadfastness in logging in and preparing all the samples for counting, making the radon calculations, maintaining the raw data sheets, and digitizing all the locations by latitude and longitude. Without the dedication and talent of Keith McCroan, the computer generated plots would have been unavailable for this paper. Without the plots much of the potential impact of the data would be lost. The author would also like to recognize the efforts of Cody Partridge and Ed Sensintaffar in developing the methods employed in sample collection and radon analysis and calibration. Mr. Partridge was also responsible for conducting the pilot study. The author especially appreciates the sincere efforts of both individuals to impart their knowledge of this project to the author. Special thanks also to the reviewers of this paper, especially to Jon Broadway and Charles Porter for their helpful suggestions, to Mardy Wilkes for typing the many drafts, and to Chuck Petko for providing editorial support. Finally, the author thanks the EPA's Office of Drinking Water for supplying computerized listings of all public groundwater systems serving 1000 or more people.

1.0 INTRODUCTION

In 1978, the Environmental Protection Agency (EPA), through its Eastern Environmental Radiation Facility (EERF), began sampling radon in drinking water. During the next two to three years, approximately 27 states were included in this pilot study, the purpose of which was to determine the need for a nationwide study of radon in drinking water; to demonstrate the feasibility of such a study; and to develop a limited data base of radon in drinking water nationwide. The subject of this paper is the nationwide study that developed from that pilot study.

The nationwide study, which began in November of 1980, was designed to systematically sample water supplies in all 48 contiguous states. The results of the study will be used, in cooperation with EPA's Office of Drinking Water (ODW), to estimate population exposures nationwide and to support future standards for radon, uranium, and other natural radioactivity in public water supplies.

The study design called for sampling only finished water; limited sampling to once per water supply; targeted composite samples or system samples instead of individual well supplies; encouraged sampling as near the source of water as possible; and excluded surface water supplies (no significant radon was detected in surface water in the pilot study) and supplies that served less than 1000 people. Our intent was to collect samples that represented what people actually consume from a given public water supply.

Of the more than 2500 public water supplies that we sampled, more than 95 percent met our criteria. Only about one percent were surface systems (less than 30 supplies), and less than four percent of the groundwater supplies served populations of less than 1000.

The scope of this study is also noteworthy. Although we sampled only about five percent of the total number of groundwater supplies in the 48 contiguous states of the U.S., those samples represent 45 percent of the water consumed by U.S. groundwater users.

2.0 SAMPLING AND ANALYSIS METHODS

2.1 Sampling Method

The sampling procedure for radon in drinking water is described in the EPA/EERF manual (Ref. 1). This method is reliable if the instructions outlined in the manual are followed carefully.

Two samples per water supply are taken. Collecting and analyzing two samples provides us a backup sample if one vial is broken or leaks during shipment, gives us a measure of overall precision in sampling and measuring radon, and permits the computation of an average value instead of a single value.

After collection, the 10 ml aliquot of sampled water is added to a 20 ml glass scintillation vial containing 10 ml of mineral oil based liquid scintillator. The two scintillation vials are carefully packed in a mailing tube to be shipped to the EERF. The vials are separated from each other with newspaper, a paper towel, or other packing material and must be well packed to withstand shipping impacts. A completed sampling and analysis form, which identifies the samples and provides information necessary for calculating radon concentration, is returned with the vials. The vials must be mailed on the collection day or the following day to avoid unnecessary radon decay. Radon in samples received seven to ten days after collection has generally decayed beyond detection, unless the sample has a relatively large initial radon concentration (e.g., 1000 pCi/l or greater).

2.2 Liquid Scintillation Counting

Radon samples are analyzed by liquid scintillation counting. Our method varies from the Prichard and Gesell procedure (Ref. 2) in that we use a mineral oil based scintillation mixture instead of a toluene-based liquid scintillation fluid. We use the mineral oil mixture because it has a higher flash point, which allows sample shipment through regular mail without restrictions. We also use 10 ml of mix instead of 5 ml. Typical instrument settings are 1.0 for the gain with a wide open window for the energy discriminator (i.e., lower level 0.1 and upper level 10.0). Background count rate varies from about 35 counts per minute (cpm) to 45 cpm depending on the instrument. A 50 minute count gives a minimum detectable level (MDL) of about 15 pCi/l, while a 20 minute count raises the MDL to about 23 pCi/l. The MDL is a function of the counting efficiency, counting time, and background count rate (Ref. 3 and Ref. 4).

2.3 Ra-226 Calibration

A traceable National Bureau of Standards Ra-226 standard solution is used in calibration. A known quantity of Ra-226 is added to a known volume of distilled water; 10 ml aliquot of the Ra-226 solution is combined with 10 ml of a mineral oil based scintillator mix and sealed in a 20 ml glass scintillation vial; and radon, the daughter of Ra-226, is allowed to build up for approximately 21 days. At this point, radon, for all practical purposes, has reached secular equilibrium with Ra-226. By shaking the vial, nearly all the radon is transferred to the scintillator phase (radon is highly soluble in the scintillator). By waiting three hours before counting, the radon short-lived daughters are allowed to build up to secular equilibrium with radon. The Ra-226 remains in the aqueous phase and, therefore, does not contribute significantly to the count rate. This was verified by counting the standard before significant buildup of radon occurred. A near background count rate was observed. The slight increase in count rate would be due to the Ra-226 at the aqueous/scintillator interface. The standard and the background samples are counted for 50 minutes or longer. To obtain the cpm/pCi conversion factor, the background cpm is subtracted from the gross cpm for the standard and the difference is divided by the known radon activity in pCi. The radon activity equals the Ra-226 activity at secular equilibrium. A typical cpm/pCi conversion factor is about 10.2 cpm/pCi of radon. This relates to a counting efficiency of about 90%.

2.4 Rn-222 Concentration Determination

The radon concentration in pCi/l in the sample is given by--

pCi/l = (net cpm/c.f./decay) x (1000 ml/liter/10 ml),

where

net cpm = gross cpm - background cpm,

c.f. = cpm/pCi conversion factor,

decay = exp(-7.56E-3 x time), and

time = time lapse from time of collection to time of counting in hours.

Two sigma counting error in percent = 2 x (gross cpm/length of count + background/length of count)(1/2 power)/net cpm x 100.

2.5 Precision and Accuracy of Rn-222 Determinations

Using 246 duplicate pairs of data (where concentrations ranged from 100-500,000 pCi/l) collected from November 1978 through February 1981, a plot of the average range between paired data versus the average concentration using least squares linear regression analysis is shown in Figure 1. The slope of the line indicates an approximate five percent degree of precision over the entire range of concentrations (100-500,000 pCi/l). Note that the highest concentration interval is not plotted in Figure 1, but it was used to calculate the least squares fit.

The EERF participated in an intercomparison study with the University of Texas School of Public Health at Houston where several different methods were employed (Ref. 5). Our data compared favorably with the overall mean values obtained by the study and control sample results as shown in Table 1.

An informal cross-check with the University of South Carolina (USC) Geology Department was also conducted. A set of samples was collected from ten different water supplies (0-7000 pCi/l) which had a wide range of radon concentrations. The results of this intercomparison are shown in Figure 2. A very high correlation is seen between the two sets of data. The USC data were obtained by an entirely different method of collection and analysis. In the USC method, a large volume sample is collected followed by radon de-emanation and alpha counting.

2.6 Other Radionuclide Determinations

In addition to the radon samples, a one gallon cubitainer water sample was collected for each water supply included in the study. These samples allowed us to obtain other data on natural radioactivity in public drinking water for very little extra collection effort. Our analyses of these samples were guided, generally, by the requirements of the Safe Drinking Water Act (SDWA) (Ref. 6).

All samples were analyzed for gross alpha and gross beta. If the gross alpha was equal to or greater than 5 pCi/l, a Ra-226 analysis was performed. Shortly after the study began, the cutoff for Ra-226 analyses was dropped to 3 pCi/l to provide more data. Ra-228 analyses were performed for samples where the Ra-226 was equal to or greater than 3 pCi/l. During the second half of the study, samples whose gross beta exceeded 15 pCi/l were also analyzed for Ra-228. Where the gross

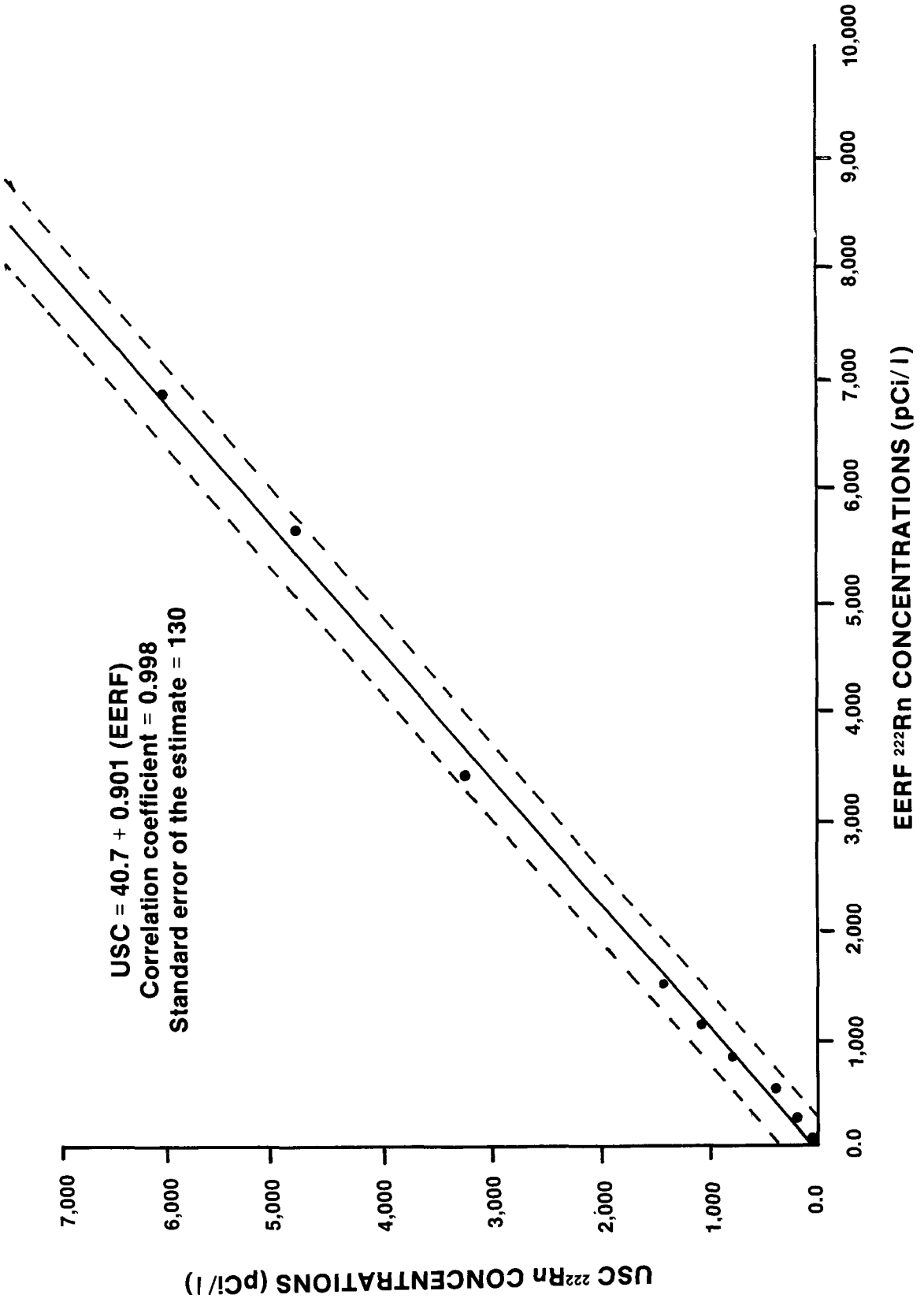

Fig. 2. The results of a ^{222}Rn cross-check.

time. The vast majority of the samples were analyzed under the lowest cutoff criterion. After performing more than 100 thorium analyses, it was decided that thorium analyses were unnecessary because the concentrations found in groundwater were very low (typically less than 0.1 pCi/l for Th-227, Th-228, Th-230, and Th-232). At these levels, the measurements have an inherent uncertainty. One Ra-226, uranium, and thorium analysis was performed for each state involved in the study, regardless of whether any sample from a given state met our cutoff criteria. This provided us baseline data for each state.

3.0 RESULTS

Locations of the more than 2500 public water supplies sampled in this study are shown in Figure 3. Nationwide, the public groundwater systems represent about 45 percent of the total groundwater usage or about five percent of the total number of public groundwater systems. Thirteen states were not included in the study, primarily because of a shortage of manpower and money. Even though the state health departments were reimbursed for collecting the samples, the reimbursement did not cover actual expenses incurred in the collection effort.

Results for radon, gross alpha, gross beta, Ra-226, Ra-228, total radium, and total uranium are summarized in Table 2. Buildup of radon and its short-lived daughters during typical household activities involving water usage can cause a significant indoor working level (WL). Based on the model used by Partridge et al. (Ref. 8), a 0.01 WL is possible for a radon in water concentration of 10,000 pCi/l and a relatively slow turnover rate of air (e.g., 0.25 air changes per hour). Energy efficient homes can have a ventilation rate somewhat less than 0.25 air changes per hour resulting in a higher WL.

Figures 4 and 5 present nationwide radon concentrations in public water supplies. Elevated radon levels are seen in the New England states, North and South Carolina, Georgia, Virginia, and western states such as Arizona, Colorado, Nevada, Montana, and Wyoming. Actual individual sample radon concentrations range from essentially zero to greater than 16,000 pCi/l. Some localized averaging is used to generate the plots. The purpose of the plots is to show general trends nationwide for inclusion in this paper. Actual individual radon concentrations for a given water supply are available and will be used in the final analysis of the data. These comments also apply to other natural radioactivity results presented in this paper.

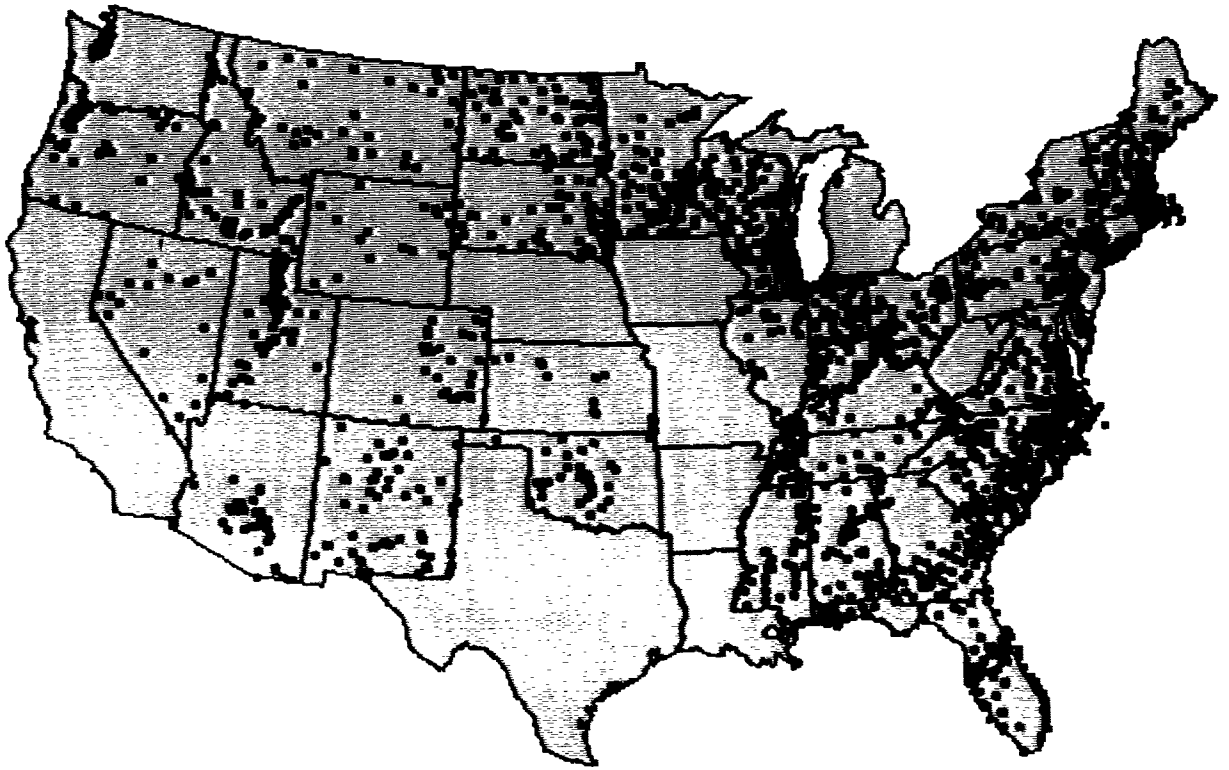

Fig. 3. Locations of public water supply samples collected 1981-1982.

TABLE 2

SUMMARY OF RADON AND OTHER NATURAL RADIOACTIVITY RESULTS

Concentration Range, pCi/l	Approximate Number of Public Water Supplies	
Radon		
> 10,000	3	(0.1%)*
5,000-10,000	7	(0.3%)
1,000- 5,000	160	(6 %)
500- 1,000	250	(10 %)
< 500	2100	(83 %)
Arithmetic mean = 340 pCi/l		
Geometric mean = 100 pCi/l		
Gross Alpha		
> 15	51	(2 %)
10 -15	56	(2 %)
5 -10	128	(5 %)
< 5	2300	(91 %)
Arithmetic mean = 1.8 pCi/l		
Geometric mean = 0.6 pCi/l		
Gross Beta		
> 15	60	(2 %)
10 -15	75	(3 %)
5 -10	340	(14 %)
< 5	2040	(81 %)
Arithmetic mean = 3.4 pCi/l		
Geometric mean = 2.1 pCi/l		
Ra-226		
> 5	34	(9 %)
2 - 5	85	(22 %)
1 - 2	35	(9 %)
< 1	232	(60 %)
Arithmetic mean = 1.6 pCi/l		
Geometric mean = 0.6 pCi/l		

(TABLE 2)-Continued

Concentration Range, pCi/l	Approximate Number of Public Water Supplies	
Ra-228		
> 5	25	(25 %)
2 - 5	41	(41 %)
1 - 2	12	(12 %)
< 1	22	(22 %)
Arithmetic mean = 3.5 pCi/l		
Geometric mean = 2.3 pCi/l		
Total Ra		
> 10	29	(32 %)
5 -10	43	(47 %)
2 - 5	18	(20 %)
1 - 2	1	(1 %)
< 1	0	(0 %)
Arithmetic mean = 8.5 pCi/l		
Geometric mean = 7.6 pCi/l		
Total U		
> 20	18	(5 %)
10 -20	31	(9 %)
5 -10	51	(15 %)
2 - 5	96	(28 %)
< 2	148	(43 %)
Arithmetic mean = 5.1 pCi/l		
Geometric mean = 1.9 pCi/l		

* Percent of total

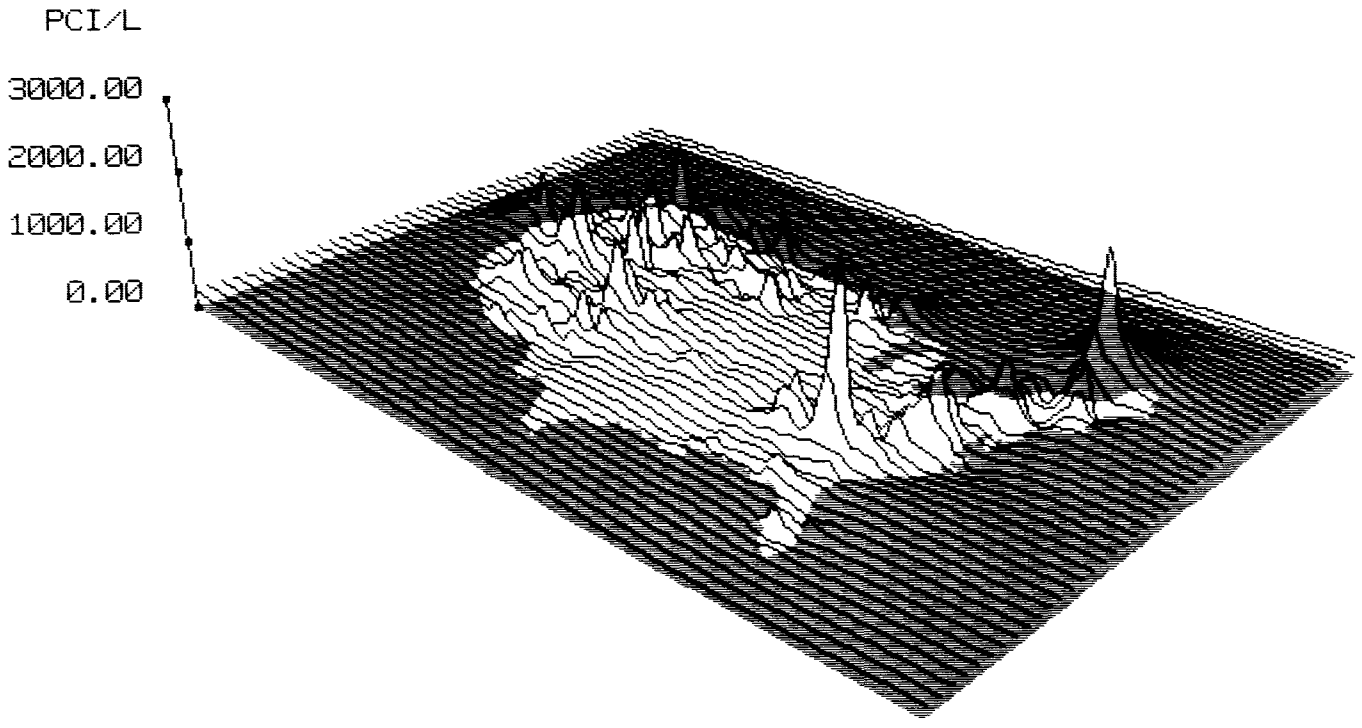

Fig. 4. Average Rn-222 concentrations in drinking water samples collected 1981-1982.

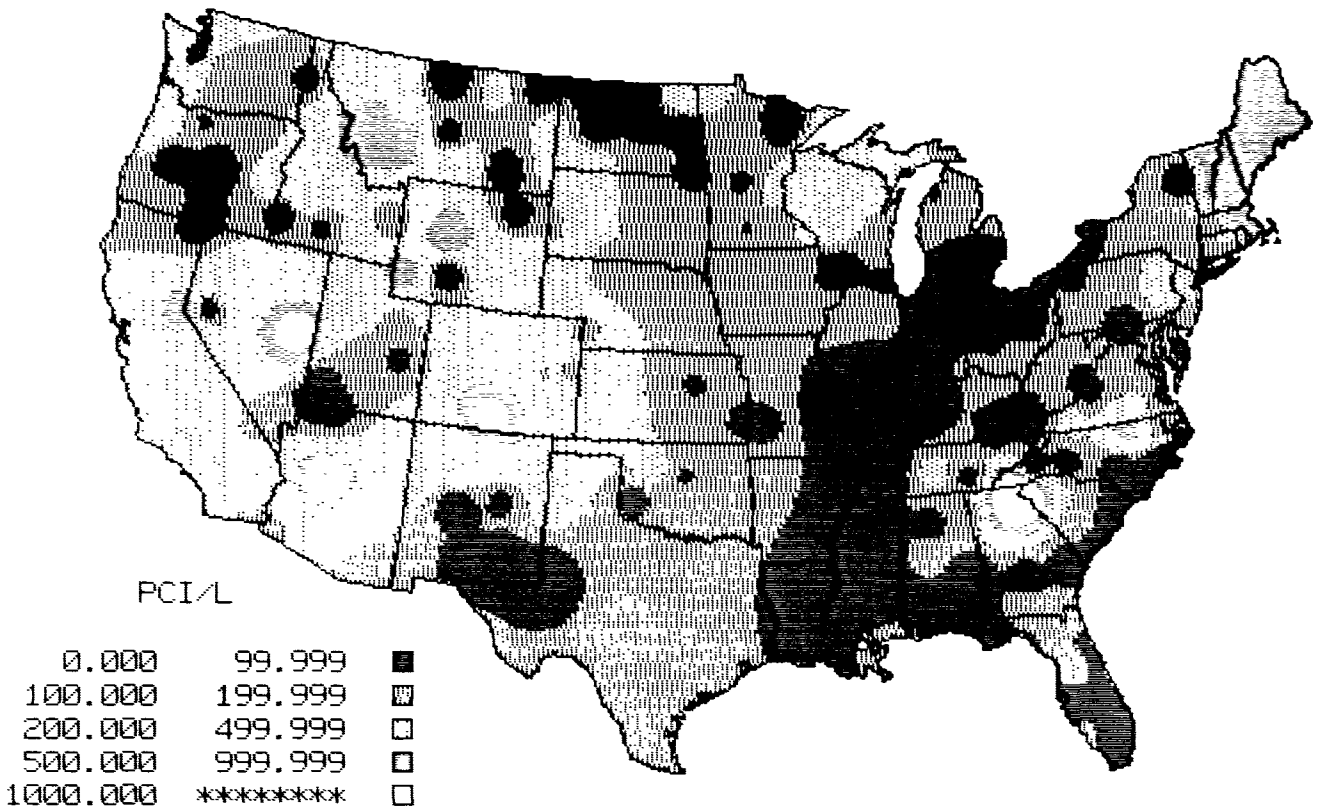

Fig. 5. Average Rn-222 concentrations in drinking water samples collected 1981-1982.

Table 3 lists the means for seven states where radon results were not available from our nationwide study. These means are based on results from our pilot study and Prichard's study (Ref. 7). By comparing the geometric means of Table 3 for individual states with Figures 4 and 5, it is evident that the radon results used to generate these figures, which do not include the results of Table 3, do show a reasonably good approximation for those seven states. Having radon results from all 48 contiguous states would be desirable, but in practice may not be necessary.

TABLE 3

ARITHMETIC AND GEOMETRIC MEANS FOR MISSING STATES

State	Radon Concentration, pCi/l Arithmetic Mean (Geometric Mean)	Approximate Number of Public Water Supplies
AR	120 (12)	22
CA	1200(470)	9
IA	1500(220)	40
LA*	180 (55)	15
MO	300 (24)	69
NE*	320(200)	16
NJ	690(300)	19
US	340(100)	2500

* Ref. 7; results for all other states are from our pilot study.

A regional map of the New England states is shown in Figure 6. An area of elevated radon concentration is seen stretching from northern Vermont and New Hampshire through Massachusetts into Rhode Island. All locations were used in producing this map, i.e., no localized averaging was employed. As can be seen, more detail is preserved in this regional map versus the U.S. map.

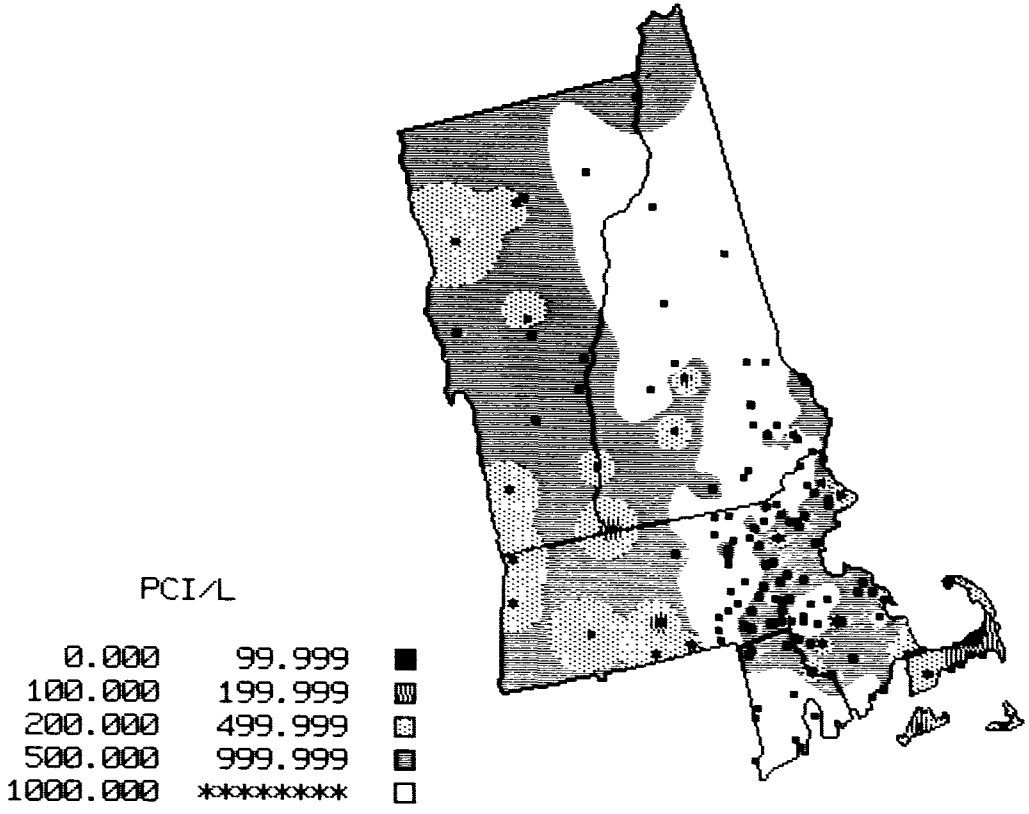

Fig. 6. Average Rn-222 concentrations in drinking water in Massachusetts, New Hampshire, Rhode Island, and Vermont, 1981-1982.

Nationwide gross alpha concentrations are displayed in Figure 7. Most of the U.S. is below 3 pCi/l. In general, only those locations with elevated uranium show elevated levels.

Ra-226 concentrations are shown in Figures 8 and 9. North and South Carolina, Georgia, Florida, and the midwest states of Illinois, Wisconsin, Minnesota, and Kansas show elevated levels. Almost no Ra-226 is seen in the western states.

Total uranium (U-234 plus U-238) concentrations are presented in Figures 10 and 11. The western states of New Mexico, Colorado, Wyoming, and Montana and a few eastern states such as Maine and Pennsylvania exhibit elevated levels.

The ratio of U-234 to U-238 is of interest to those areas where elevated total uranium concentrations exist. This interest stems from the fact that a lower initial cost and simpler method of analyzing for total uranium (fluorometric method versus alpha spectroscopy used at the EERF) assumes secular equilibrium between U-234 and U-238 while the fluorometric method only measures the U-238 content. In certain cases the total uranium activity may be severely underestimated using the fluorometric method.

Using uranium results whose total uranium exceeds 3.5 pCi/l, Figures 12 and 13 were generated. Nearly all the ratios are between one and two with an arithmetic mean of 1.8 and a geometric mean of 1.7. Low activity samples with their inherent uncertainty are not included.

Some of the water supplies sampled during the nationwide study did not meet our sampling criteria. These include surface water supplies and groundwater supplies serving less than 1000 people. A decision was made to include these supplies in the analysis of results, since they are public water systems.

The natural radioactivity associated with the surface supplies is very low. This includes radon, gross alpha, and gross beta. The groundwater supplies serving less than 1000 people did not significantly alter the overall results. In general, the same concentration pattern is observed in the less than 1000 people groundwater supplies as is observed in the greater than 1000 people systems.

4.0 SUMMARY

Samples from more than 2500 public water supplies representing 35 states were collected. For the most part, samples were from public groundwater supplies serving 1000 or more people. Although we sampled only about five percent of the total number of groundwater supplies in the 48 contiguous states of the U.S., those samples represent nearly 45 percent of the water consumed by the U.S. groundwater users in the 48 contiguous states. Our intent was to collect samples that represented what people actually consume from a given public groundwater supply.

The arithmetic means for radon, Ra-226, and total uranium were calculated to be 340 pCi/l, 1.6 pCi/l, and 5.1 pCi/l, respectively. The corresponding geometric means for radon, Ra-226, and total uranium were found to be 100 pCi/l, 0.6 pCi/l, and 1.9 pCi/l, respectively.

The arithmetic mean for the U-234/U-238 ratio was determined to be 1.8 for higher activity samples (total uranium exceeded 3.5 pCi/l), while the corresponding geometric mean was 1.7. In most cases, total uranium activity determined by the fluorometric method will not be significantly underestimated if a correction factor is applied based on the mean U-234/U-238 ratio for a given geographic region or possibly the entire U.S.

The results of this nationwide study will be used, in cooperation with EPA's Office of Drinking Water, to estimate population exposures nationwide and to support future standards for radon, uranium, and other natural radioactivity in public water supplies.

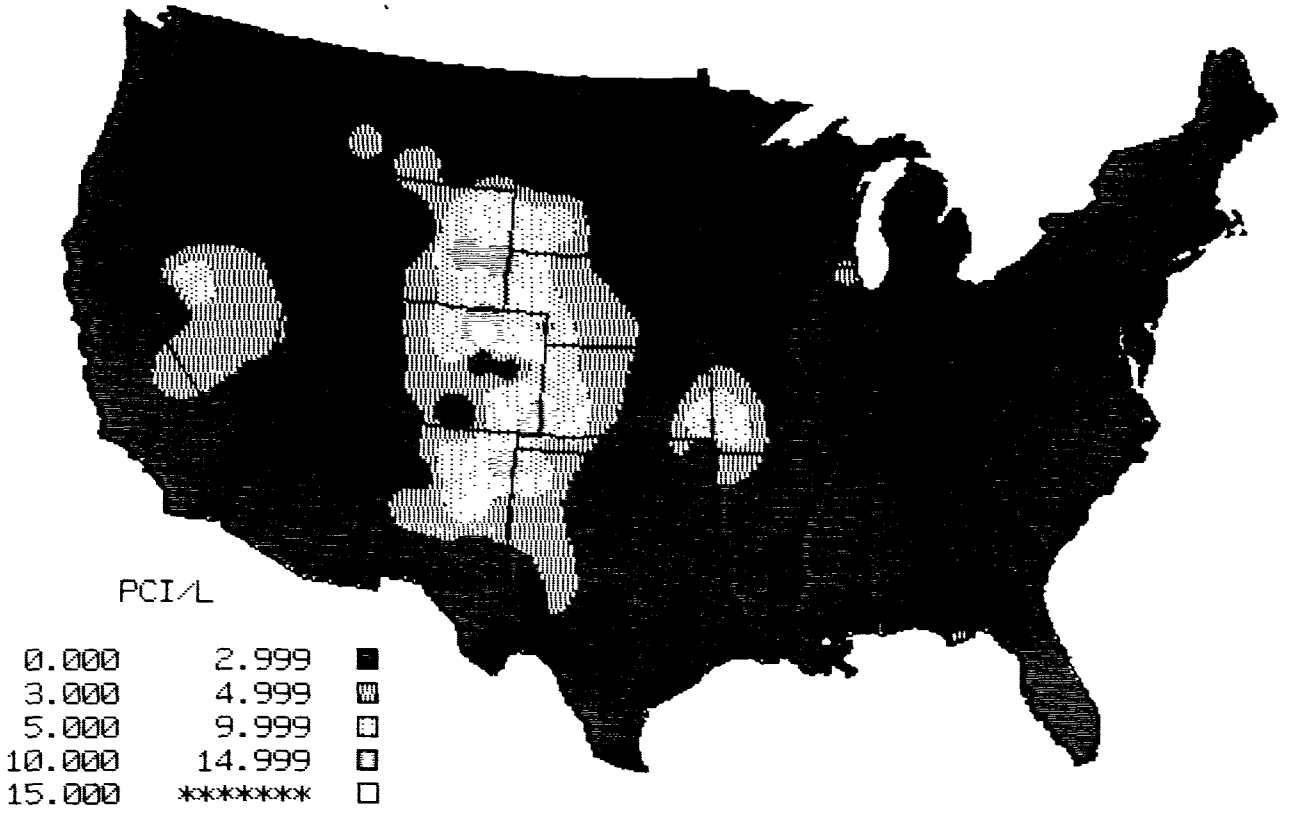

Fig. 7. Average gross alpha concentrations in drinking water samples collected 1981-1982.

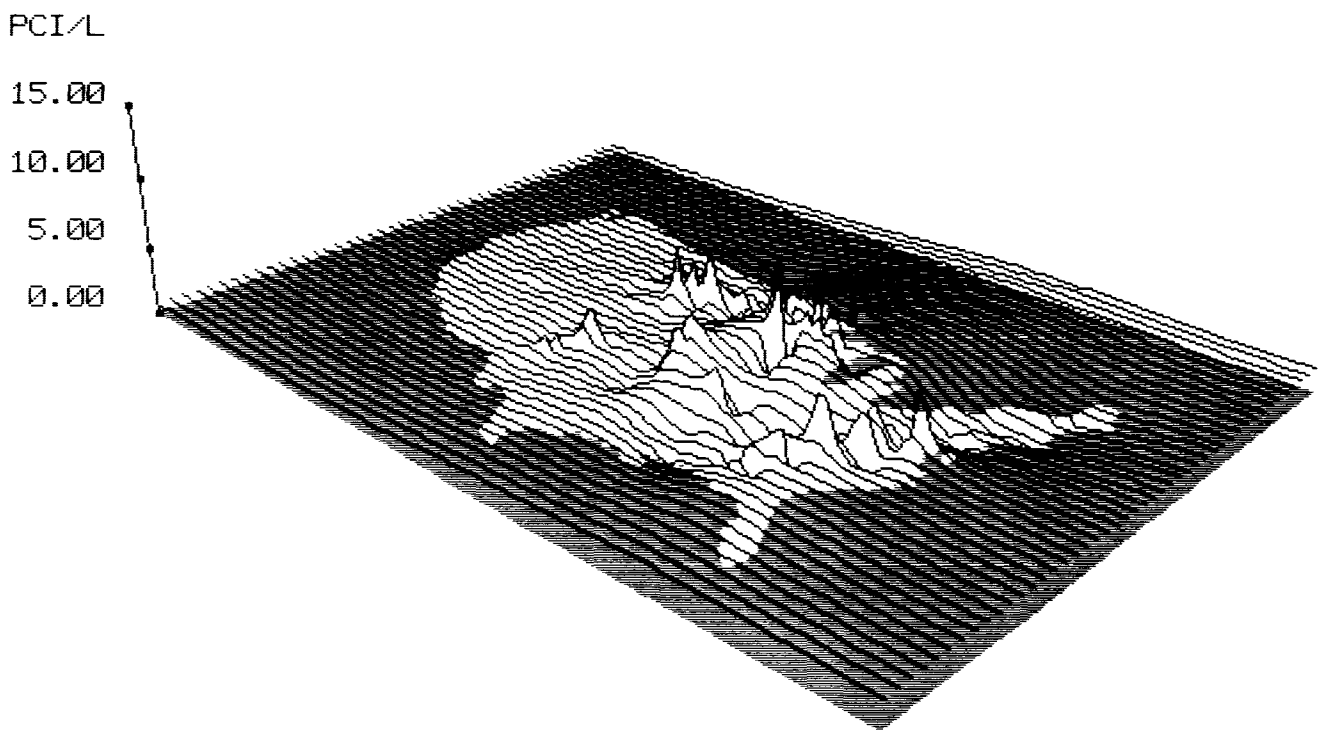

Fig. 8. Average Ra-226 concentrations in drinking water samples collected 1981-1982.

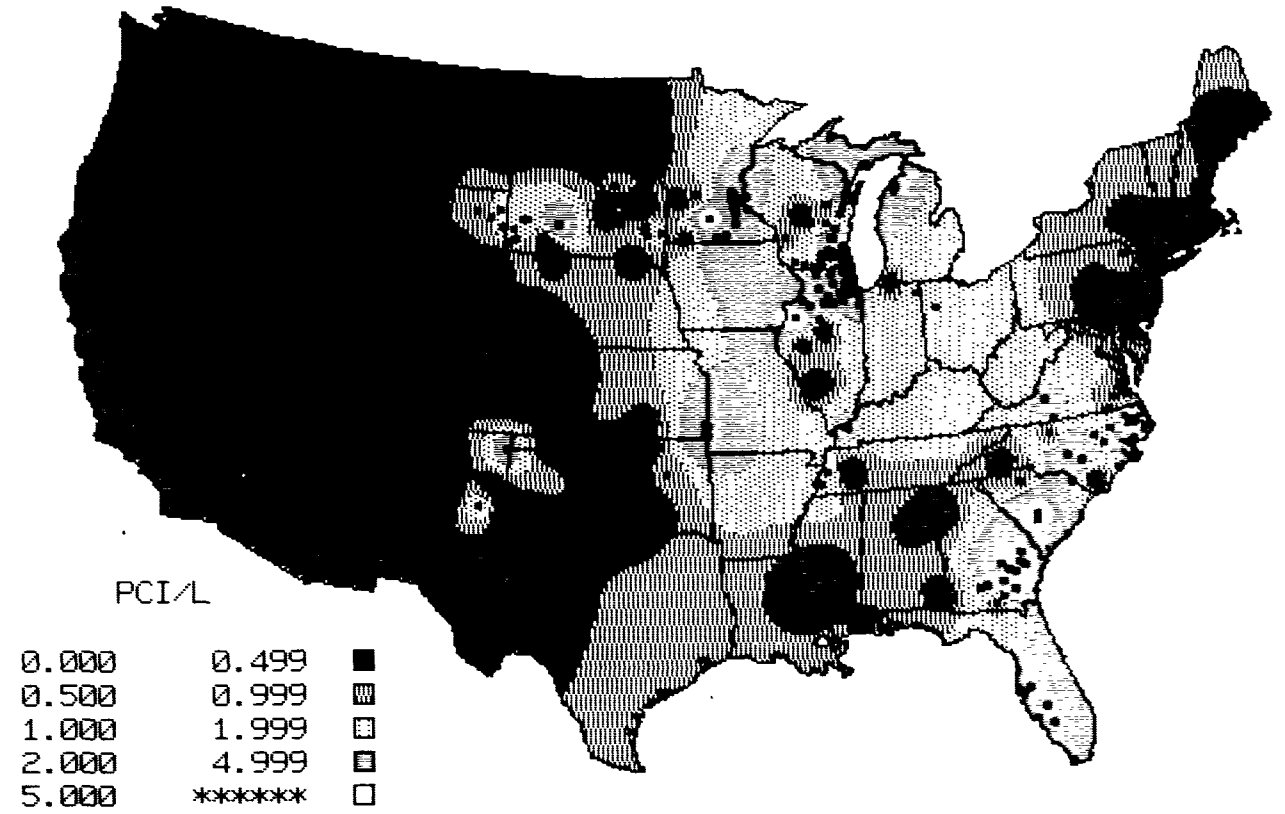

Fig. 9. Average Ra-226 concentrations in drinking water samples collected 1981-1982.

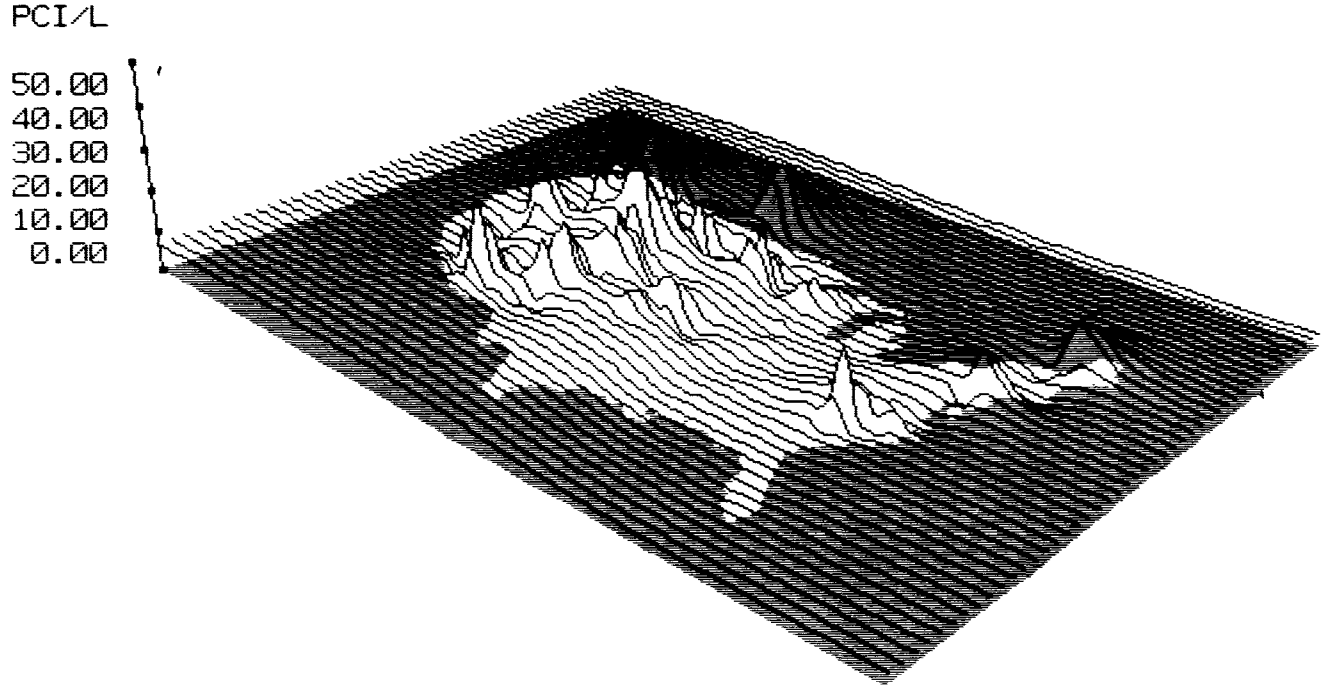

Fig. 10. Average total uranium concentrations in drinking water samples collected 1981-1982.

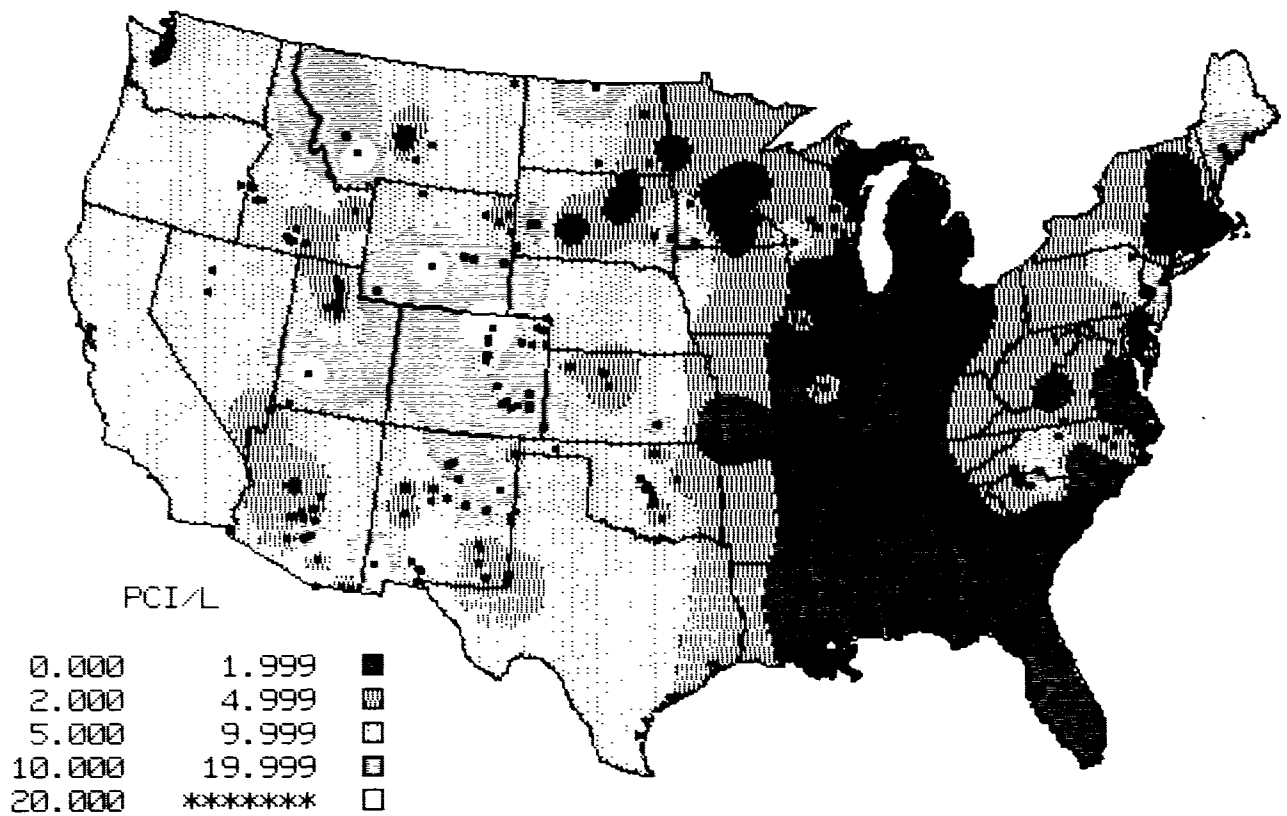

Fig. 11. Average total uranium concentrations in drinking water samples collected 1981-1982.

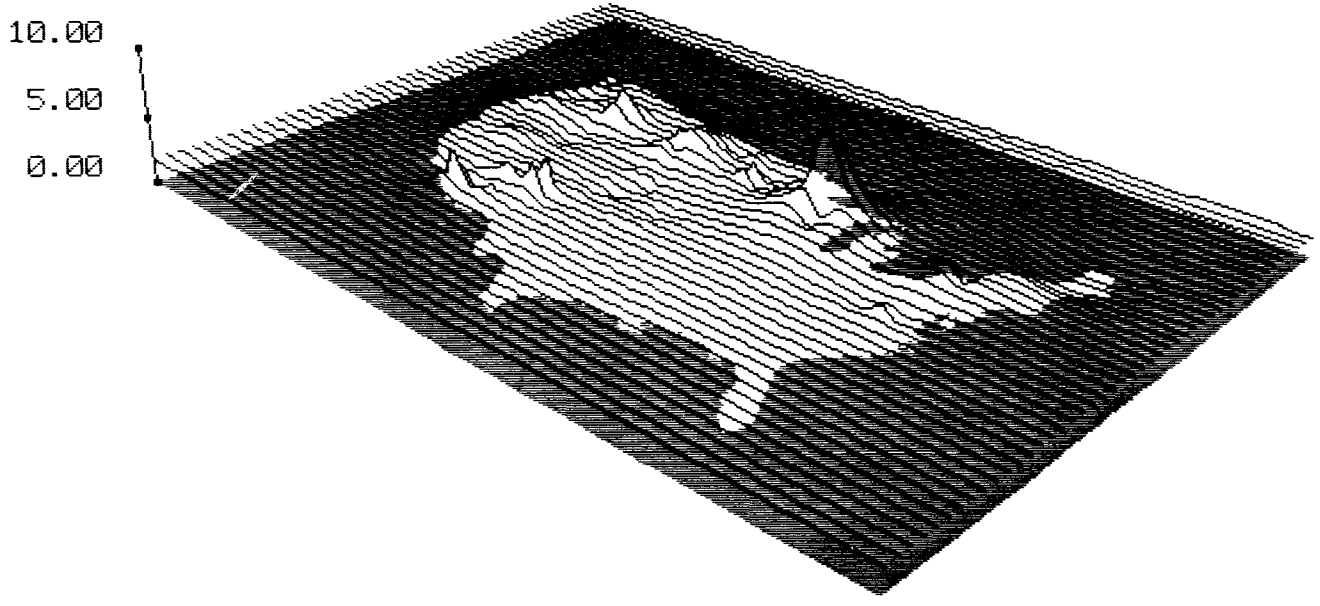

Fig. 12. U-234/U-238 ratios in drinking water samples (samples with total uranium > 3.5 pCi/l) collected 1981-1982.

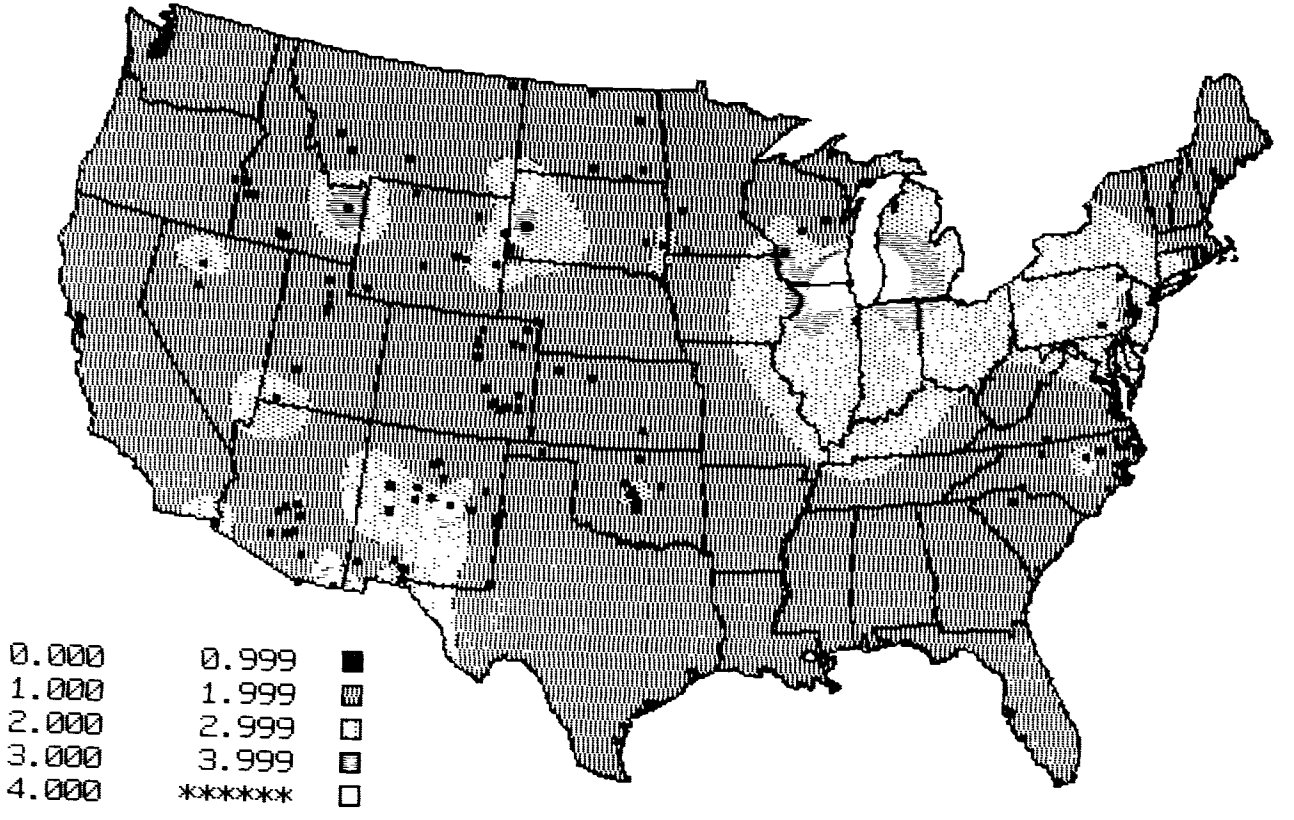

Fig. 13. U-234/U-238 ratios in drinking water samples (samples with total uranium > 3.5 pCi/l) collected 1981-1982.

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