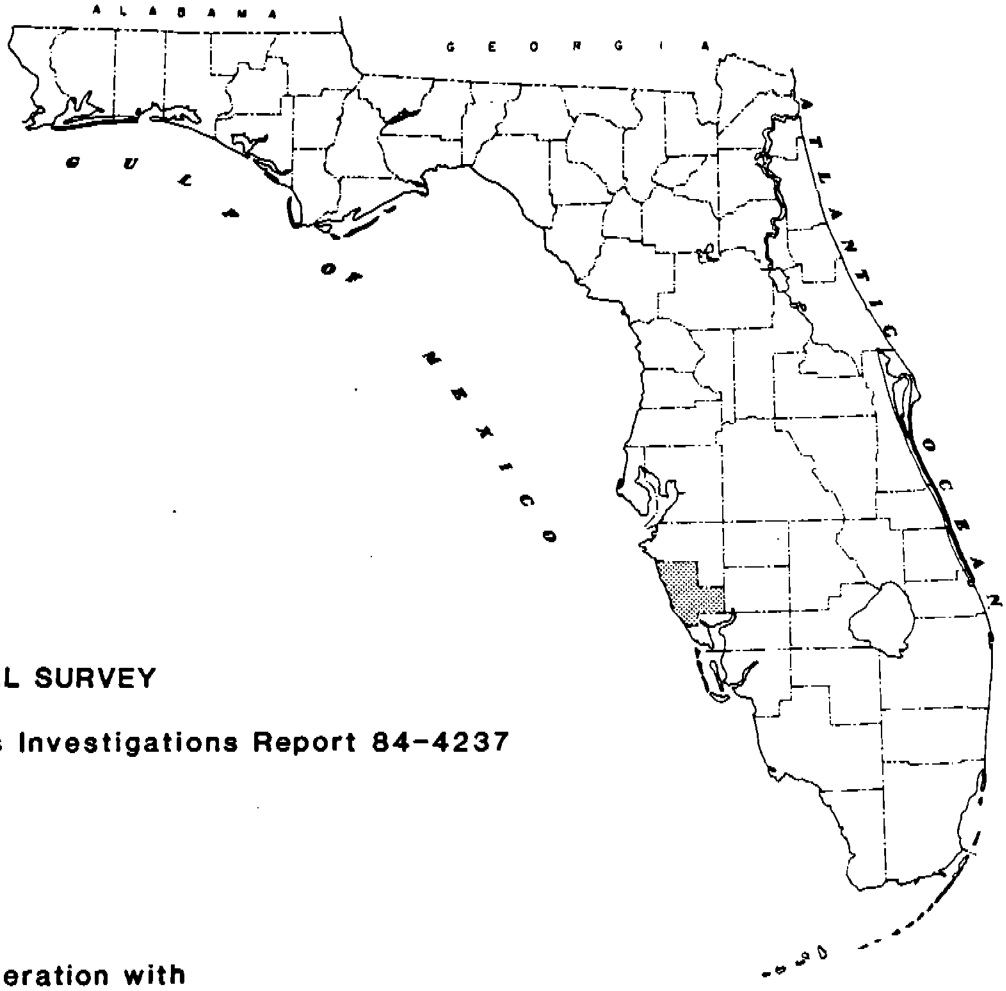
84-4237 OCCURRENCE OF NATURAL RADIUM-226 RADIOACTIVITY IN GROUND WATER OF SARASOTA COUNTY, FLORIDA 84-4237

OCCURRENCE OF NATURAL RADIUM-226 RADIOACTIVITY IN GROUND WATER OF SARASOTA COUNTY, FLORIDA



U.S. GEOLOGICAL SURVEY

Water-Resources Investigations Report 84-4237

Prepared in cooperation with

SARASOTA COUNTY, FLORIDA

OCCURRENCE OF NATURAL RADIUM-226 RADIOACTIVITY IN GROUND WATER OF SARASOTA COUNTY, FLORIDA

By Ronald L. Miller and H. Sutcliffe, Jr.

U.S. GEOLOGICAL SURVEY

Water-Resources Investigations Report 84-4237

Prepared in cooperation with

SARASOTA COUNTY, FLORIDA

Tallahassee, Florida

1985

UNITED STATES DEPARTMENT OF THE INTERIOR

WILLIAM P. CLARK, Secretary

GEOLOGICAL SURVEY

Dallas L. Peck, Director

For additional information write to:

District Chief
U.S. Geological Survey
Suite 3015
227 North Bronough Street
Tallahassee, Florida 32301

Copies of this report can be purchased from:

Open-File Services Section
Western Distribution Branch
U.S. Geological Survey
Box 25425, Federal Center
Denver, Colorado 80225
(Telephone: (303) 236-7476)

CONTENTS

ILLUSTRATIONS

TABLES

GLOSSARY

Activity (thermodynamic) - an effective concentration of a chemical species. Measured (analytical) concentrations of ions in solution are adjusted for nonideal behavior so that equations that assume ideal behavior can be used. Some form of the Debye-Huckel equation is generally used to calculate an activity coefficient that is multiplied by the analytical concentration to get the activity of an ion. If complexation is significant, a correction must be applied for it.

Curie - a curie is that quantity of any radioisotope that undergoes 3.7×10^{10} disintegrations per second (Weist, 1976).

Distribution factor - a term that considers the effect of uneven distributions of internally deposited radionuclides in biological systems, such as the concentration of radium in bones (U.S. Atomic Energy Commission, 1967).

Dose equivalent - the amount of effective radiation in rems when modifying factors are applied. It is the product of the absorbed dose multiplied by a quality factor multiplied by a distribution factor. The dose equivalent is a dose that has been adjusted for the location of the source from an organ and for the difference in damage done by different kinds of radiation (U.S. Atomic Energy Commission, 1967).

Factor analysis - a type of data analysis that uses correlations between variables of a set of data to reduce the original number of variables to the minimal number of new variables, called factors, that contain the same amount of information. The new variables are linear combinations of the original variables and can provide insight into logical interpretations of large numbers of data that might otherwise be difficult to analyze.

Millirem - one thousandth of a rem.

Rad - acronym for radiation absorbed dose. It is the basic unit for measuring absorbed doses of ionizing radiation. A rad means that 100 ergs of radiation energy was absorbed per gram of absorbing material (U.S. Atomic Energy Commission, 1967).

Radioactivity - intensity of radiation. The unit used in this report is the picocurie, which is 1×10^{-12} curies. A picocurie corresponds to 2.220 radioactive disintegrations per minute. Radioactivity is often shortened to activity, but the use of activity for radioactivity was avoided in this report so that it would not be confused with thermodynamic (chemical) activity.

Relative biological effectiveness (RBE) - a factor used to compare the effectiveness of different types of ionizing radiation to produce damage. Usually X-rays are the reference type of radiation. The RBE is one for beta, gamma, and X-radiation and for conversion electrons (orbital electrons emitted when the nucleus drops to a lower energy state). The RBE is 1.7 for beta emitters with energies less than or equal to 0.03 million electron volts. The RBE is 10 for alpha particles and 20 for recoil nuclei (U.S. Atomic Energy Commission, 1967).

Rem - acronym for roentgen equivalent man. The unit dose of any ionizing radiation that produces the same biological effect (damage) as a unit of absorbed dose of ordinary X-rays. The dose in rems is equal to absorbed dose in rads multiplied by the relative biological effectiveness (U.S. Atomic Energy Commission, 1967).

Roentgen - a unit of exposure to ionizing radiation (X-ray, or particles with sufficient energy to ionize a substance through which it passes). It is that amount of gamma- or X-rays required to produce ions carrying one electrostatic unit of electrical charge in one cubic centimeter of dry air under standard conditions (U.S. Atomic Energy Commission, 1967).

Standard conditions - Standard conditions for gases are zero degrees Celsius and one atmosphere of pressure. One atmosphere of pressure is also described as 760 millimeters of mercury or 101325 pascal (Weast, 1976).

OCCURRENCE OF NATURAL RADIUM-226 RADIOACTIVITY IN GROUND WATER OF SARASOTA COUNTY, FLORIDA

By Ronald L. Miller and H. Sutcliffe, Jr.

ABSTRACT

Water that contains radium-226 radioactivity in excess of the 5.0-picocurie-per-liter limit set for radium-226 plus radium-228 in the National Interim Primary Drinking Water Regulations was obtained from the majority of wells sampled throughout Sarasota County. A comparison of data from different aquifers showed that greater radium-226 radioactivities occurred in the intermediate aquifer where phosphate pebbles occur in a semiconsolidated matrix than occurred in deeper aquifers. The highest radioactivity determined for radium-226 was 110 picocuries per liter in a saline water sample.

Analysis of data suggests that a major fraction of radium-226 is released by alpha-particle recoil of thorium-230 or its precursors. Mineralized water competes with radium-226 for ion exchange and sorption sites and consequently causes elevated concentrations of dissolved radium-226. Two types of mineralized water are present in Sarasota County. One type is a marine-like water, presumably associated with saltwater encroachment in coastal areas; the other is a calcium magnesium strontium sulfate bicarbonate type. The hypothesis of alpha-particle recoil and competition for ion exchange and sorption sites would also explain the lower radium-226 radioactivities in the present phosphate mining areas, such as Polk County. Water in contact with phosphate ores in Polk County usually contains less dissolved solids than in Sarasota County. In addition, dissolution of aquifer materials that contain radium-226 and its precursors will also occur. High radium-226 radioactivities could also occur in the presence of mineralized water because activity coefficients are lower in mineralized water and the solubility of some minerals is greater.

Water that contains the highest concentrations of radium-226 usually contains enough water hardness or dissolved solids that it would not be used for domestic purposes without treatment. Thus, ion exchange softening to reduce hardness or reverse osmosis to reduce dissolved-solids concentrations will also reduce radium-226 radioactivities to less than the 5.0-picocurie-per-liter limit for drinking water.

INTRODUCTION

Analyses of water from wells in Sarasota County have indicated that some ground water contains radionuclides, particularly radium-226, in concentrations that exceed recommended limits for radium-226 plus radium-228 in public water supplies (U.S. Environmental Protection Agency, 1976). Past sampling, however, has been sparse, unsystematic, and from wells that were open to more than one water-bearing zone. Thus, little was known of the vertical distribution of radionuclides in ground water or hydrogeologic controls that determine their distribution.

Interest in radioactivity in Sarasota County has increased in recent years because of concern about the effects of proposed phosphate mining on concentrations of radionuclides. Companies are currently mining phosphate in Manatee County, which borders Sarasota County to the north, and are interested in mining in De Soto County, which borders Sarasota County to the east.

Radium-226 is only one of many radionuclides present in the natural environment. Radium-226 was investigated because it was known to exceed the National Interim Primary Drinking Water Regulations of 5.0 pCi/L (picocuries per liter) for radium-226 plus radium-228 radioactivities in many ground-water samples from Sarasota County (Kaufmann and Bliss, 1977). Radium-226 has been identified as the most important naturally occurring radionuclide likely to occur in public water supplies (U.S. Environmental Protection Agency, 1976). Determinations of radium-228 radioactivities were not made during this study. Determination of parent isotopes in phosphate chemical-plant process water and phosphate ore suggests that radium-228 radioactivity would be less than 5 percent and probably about 1 percent of the radium-226 radioactivity that originates from phosphate deposits (Miller and Sutcliffe, 1982). Because radium-228 radioactivities were not available to add to the radium-226 radioactivities, the number of water samples that exceed the 5.0-pCi/L regulation for the sum of these isotopes may be higher than indicated by these data that are based on radium-226 only. If the thorium-232 series is an important source of radium, this situation will occur.

Purpose

The purpose of this report is: (1) to describe the areal and vertical distribution of dissolved radium-226 in Sarasota County, (2) to provide information for recognizing zones (during well construction) that are less likely to contain elevated concentrations of radionuclides, (3) to interpret existing knowledge of sources and behavior of radionuclides that occur in the ground water of Sarasota County, (4) to provide a basic understanding of natural radioactivity to place radium-226 in perspective, and (5) to make some general inferences about geochemical processes that may occur in phosphate mining areas in counties adjacent to Sarasota County.

Scope

Radiochemistry, geochemistry, and hydrogeology of the surficial, intermediate, and Floridan aquifer systems are discussed within the framework of public health and water-supply needs of Sarasota County. Aquifers are divided into water-bearing zones that were identified by Joyner and Sutcliffe (1976a) for the Myakka River basin that comprises most of Sarasota County. The stratigraphy of the water-bearing zones later was modified slightly by Sutcliffe and Thompson (1983) based on work in the Venice-Englewood area.

Data were collected for this study from January 1976 to July 1980. Latitude, longitude, date, time, depth, water-bearing zone, specific conductance, radium-226 radioactivity, and concentrations of major constituents are presented in tabular form in a separate data report (Sutcliffe and Miller, 1981). Water samples were collected from existing wells that were open to specific water-bearing zones or from specific water-bearing zones in new wells during drilling. One hundred ten water samples collected from discrete water-bearing zones were selected to interpret the distribution of radium-226. The locations of sampling sites are shown in figure 1. Occasional reference is made to radiochemical or hydrogeologic information for nearby counties or Florida in general.

Published data, data from the files of the U.S. Geological Survey, and information obtained from personal communications with state and local government officials were also used to interpret the distribution and behavior of radioactivity in Sarasota County.

Methods of Investigation

Existing wells in Sarasota County were inventoried to find those that were open to discrete water-bearing zones. Some public water-supply wells that are open to more than one water-bearing zone were sampled as a part of this investigation, but the data were not used to interpret the distribution or geochemistry of radium-226.

Effort was made to sample an even distribution of wells throughout the county. The Sarasota County Health Department notified U.S. Geological Survey personnel when drilling permits were issued for parts of the county where radium-226 data were lacking. In some cases, samples were collected at several depths as wells were drilled.

Wells that were drilled using the cable-tool method were chosen for sampling. This method does not require use of drilling water, drilling mud, or cement; consequently, there is a reduced chance of water samples being contaminated by drilling operations.

Water samples were collected using a bailer or, in some cases, were pumped using centrifugal or submersible pumps. Untreated samples were shipped to the U.S. Geological Survey's Denver Central Laboratory and were filtered in the laboratory. Some samples were analyzed by the radon-emanation method that is specific for radium-226 (Thatcher and others, 1977, method R-1141-76). Other samples were analyzed by the planchet-count method that includes all alpha emitting radium isotopes (Thatcher and others, 1977, method R-1142-76). Radium-228 would not be included because it is a beta emitter.

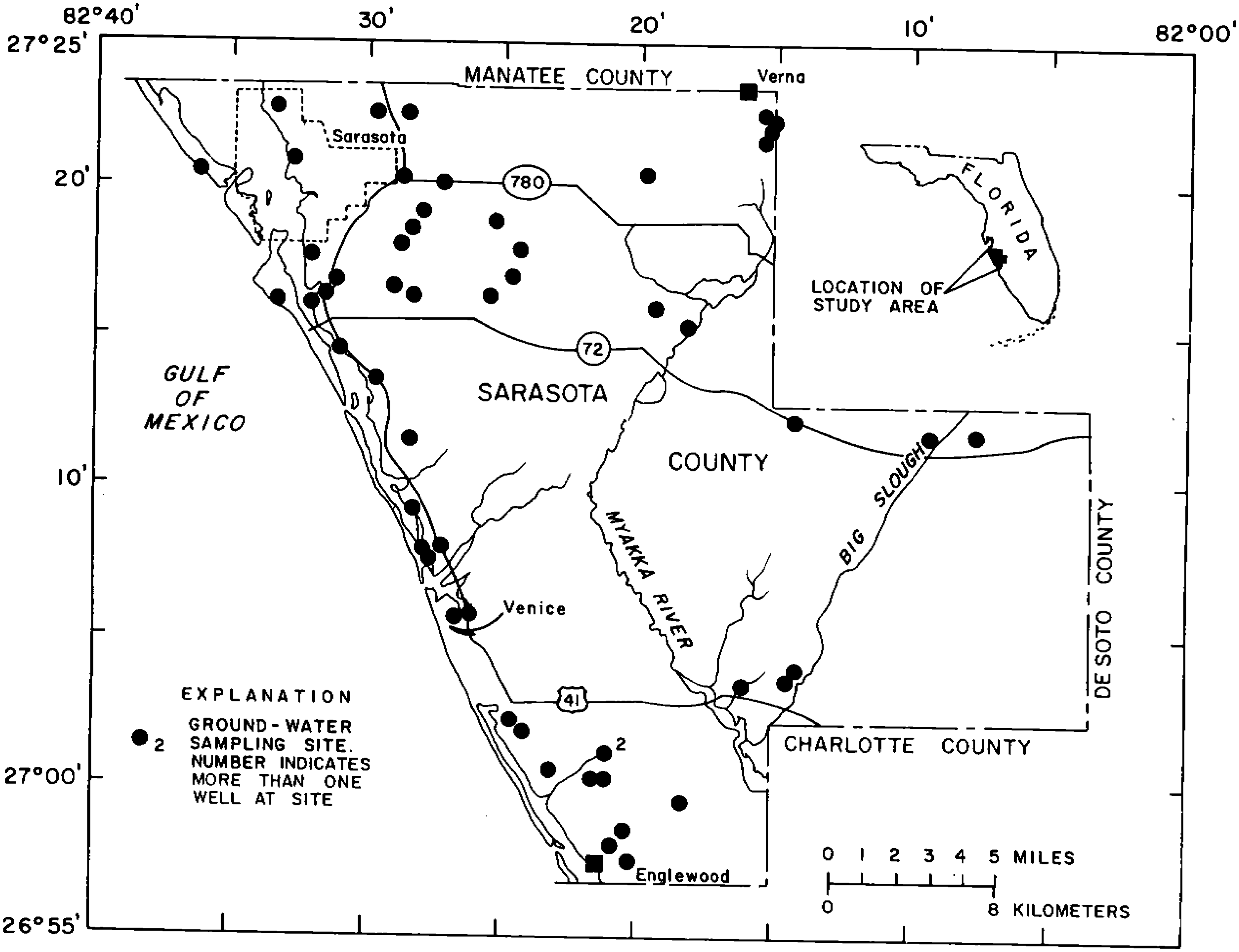


Figure 1.--Locations of study area and sampling sites.

It should be noted that additional analyses of water from wells are required to confirm that a standard is exceeded. Radioactivities in water from wells may vary with time and sampling techniques. There is an inherent measurement error in each radiochemical determination that creates some uncertainty about the exact radioactivity in a given sample.

The average ratio of dissolved-solids concentration to specific conductance for 29 ground-water samples from Sarasota County (Sutcliffe and Miller, 1981) is 0.750. This ratio was used to estimate dissolved-solids concentrations for analyses that had specific-conductance data but did not have dissolved-solids concentration data. The estimated dissolved-solids concentrations were used in combination with previously published water-quality data to define the distribution of dissolved-solids concentrations in the county.

Previous Investigations

Stringfield (1933) described the ground-water resources of Sarasota County, and Bishop (1960) described freshwater resources in the county. Joyner and Sutcliffe (1976a) studied the water resources of the Myakka River basin that comprises much of Sarasota County, and Sutcliffe and Thompson (1983) described the ground-water resources in the Venice-Englewood area. Wolansky (1983) described the hydrogeology of the Sarasota-Port Charlotte area. Kaufmann and Bliss (1977) analyzed radium-226 data using nonparametric statistics for ground water from Polk, Hardee, Hillsborough, Manatee, and De Soto Counties. They compared data from areas with and without phosphate mineralization. Sorg and others (1980) found that eight reverse-osmosis water-treatment plants in Sarasota County lowered radium-226 radioactivities below the 5.0-pCi/L drinking-water standard. Brinck and others (1978) found that most conventional water-treatment methods reduced radium-226 radioactivities in 14 water-treatment plants in Iowa and Illinois.

Acknowledgments

We acknowledge the support of the Sarasota County Commission, the County Administrator, Edgar E. Maroney, and their environmental specialist, Dr. Jeffrey L. Lincer, and the Environmental Health Section of the Sarasota County Health Department for furnishing notifications when wells would be drilled and could be sampled; the assistance of the many well drillers who collected cuttings and water samples during the drilling of wells; and particularly the Southwest Florida Water Management District, who allowed the U.S. Geological Survey to sample several core test holes and assisted in collecting and describing the cuttings that were to identify the water-bearing zones.

RADIOCHEMICAL FRAMEWORK

People are constantly exposed to low-level, natural radiation that is received from many sources. Radium-226 is but one of these sources. Some radiation is from the Earth itself and comes from radioactive decay of primordial radionuclides, which have existed since the beginning of the Earth, and their daughter radionuclides. Cosmic radiation from the sun and outer space is also a source of radiation. Another source is cosmogenic radionuclides that are produced by interaction of cosmic radiation with the Earth's atmosphere. These interactions produce unstable nuclei such as carbon-14.

The primordial radionuclides and their daughter radionuclides, including radium-226, are members of radioactive decay series, or they are nonseries radionuclides that decay in one step to stable isotopes. All elements from natural sources that have atomic numbers of 83 (bismuth) or more are radioactive and belong to the uranium-238, thorium-232, or uranium-235 series. Each series has a long-lived parent radionuclide that decays from one radionuclide to another until a stable lead isotope results. Radium-226 is a member of the uranium-238 series.

Seventeen nonseries primordial radionuclides have been identified. All except potassium-40 and rubidium-87 are scarce, long-lived radionuclides that do not contribute significantly to levels of natural radiation.

Cosmic rays are high energy electrons and nuclei of atoms from outer space (primary particles) and secondary particles produced when the primary particles interact with the Earth's atmosphere. The primary radiation from galactic sources is composed of protons (87 percent), alpha particles (11 percent), a few heavy nuclei (1 percent), and electrons (1 percent). The galactic contribution to cosmic radiation is relatively constant with time. Little solar particle radiation penetrates the Earth's magnetic field and interacts with the atmosphere except during times of solar flares (National Council on Radiation Protection and Measurements, 1975).

Cosmogenic radionuclides are continuously generated by the interaction of cosmic rays with the Earth's atmosphere or particulate matter present in the atmosphere and transported to the Earth's surface by circulation of air. Some important cosmogenic radionuclides are carbon-14, krypton-81, hydrogen-3 (tritium), beryllium-10, aluminum-26, and chlorine-36.

In addition to the naturally occurring radionuclides, manmade radionuclides are generated by nuclear tests or by activities of the nuclear industry such as electric power generation. Twenty-six such radionuclides have been identified (National Council on Radiation Protection and Measurements, 1976).

Table 1 shows the dose equivalent rates due to various natural sources of radiation to the bone marrow, gastrointestinal tract, and gonads. (See glossary for definition of dose equivalent rate.) Cosmic radiation, external terrestrial radiation, and radionuclides that have been incorporated into the body by ingestion or inhalation each contribute about one-third of the dose equivalent rate to body parts shown in table 1. Radium-226 exposure is mainly from radionuclides in the body.

Data in table 2 show average dose equivalent rates from several natural radionuclides. Ingested potassium-40 (0.00118 percent natural abundance) is an important source of natural radiation. Average data for the United States indicate that the thorium decay series contributes twice the external dose equivalent rate of the uranium decay series. This relation probably does not apply to areas with phosphate ore because of the high uranium-238 content of the ore. The thorium decay series may predominate in areas containing monazite sand (black sand), a rare earth phosphate mineral, that may occur along some parts of the coast of Sarasota County.

Table 1.--Average dose equivalent rates from natural sources of radiation in the United States

[Doses in millirems per year. G.I., gastrointestinal. Modified from National Council on Radiation Protection and Measurements (1975)]

Source	Bone marrow		G.I. tract		Gonads	
	Dose	Percent	Dose	Percent	Dose	Percent
Cosmic radiation	28	36	28	36	28	34
Cosmogenic radionuclides	0.7	1	0.7	1	0.7	1
External terrestrial	26	33	26	33	26	32
Radionuclides in the body	24	30	24	30	27	33
Rounded totals	80	100	80	100	80	100

Table 2.--Average dose equivalent rates from natural radionuclides in the United States

[Doses in millirems per year. Mode of exposure: E, external; I, internal. G.I., gastrointestinal. Modified from National Council on Radiation Protection and Measurements (1975)]

Radionuclides	Mode of exposure	Bone marrow	G.I. tract	Gonads
Carbon-14	I	0.7	0.7	0.7
Potassium-40	E	8	8	8
Potassium-40	I	15	19	19
Rubidium-87	I	0.6	0.3	0.3
Uranium series	E	6	6	6
Uranium-238 Uranium-234	I	0.9	0.8	0.8
Radium-226	I	1.2	0.2	0.2
Radon-222	I	0.4	0.4	0.4
Lead-210 Polonium-210	I	4.8	3	6
Thorium series	E	12	12	12

Radium-226 contributes about 5 percent of the exposure of bone marrow that is caused by internal radionuclides (table 2). In Sarasota County, radium-226 probably contributes a greater percentage of exposure of bone marrow caused by internal radionuclides than the national average.

For this investigation, attention was focused mainly on the uranium-238 decay series because radium-226 belongs to that series. As shown in figure 2, radium-226 is the sixth member of the uranium decay series and produces radon-222 as a daughter product when it decays. Modes of decay are shown for alpha- (α) and beta- (β) particle decay and for gamma (γ)-ray emission. The uranium-238 decay series has five minor branches as designated by the letter B where they occur in the decay series. The fraction of radionuclide that branches off is small (0.15 percent or less) in each case.

Data collected at three phosphate-industrial sites in central Florida (Miller and Sutcliffe, 1982) suggest that radon-222 will migrate more rapidly in ground water than other members of the uranium series, despite its short half-life, because of its chemical inertness. It is continuously produced by radium-226, and some small fraction of it will be released by alpha-particle recoil and will migrate away from the source of radium-226. Alpha-particle recoil could also explain the high levels of radon in confined aquifers and structures built on reclaimed land in phosphate mining areas (U.S. Environmental Protection Agency, 1975). Radon-222 and its daughters, lead-210 and polonium-210, could present health problems.

High alpha radioactivities have been measured in water from wells that are used to dewater the surficial aquifer in nearby counties that contain economically important phosphate deposits. Some alpha radioactivity may be from dissolved uranium, but much of it may be due to constant production of polonium-210 by radon-222. Alpha emitters from the thorium-232 decay series may also be involved. Radon-222 would be lost during evaporation of water and subsequent drying of residues used to measure alpha radioactivity of water samples. Radioactivities of polonium-210 as high as 200 pCi/L have been measured in ground water in Florida by J. B. Cowart (Department of Geology, Florida State University, oral commun., 1982). The presence of polonium-210 may be responsible for the occurrence of alpha radioactivities that are frequently in excess of the radioactivities of radium-226 and uranium isotopes. Carbon-14, potassium-40, and rubidium-87 are not alpha emitters, so members of the uranium and thorium decay series are probably the predominant alpha emitters. If this is the case, the loss of radon-222 gas and the length of time between sample collection and determination of alpha radioactivity would have a significant impact on the measured alpha radioactivity due to rapid decay of short-lived daughters of radon-222, namely polonium-218 and polonium-214.

Table 3 shows dissolved alpha, beta, radium-226, and uranium radioactivities (all isotopes) for selected surface-water and ground-water sites in southwestern Florida (some outside study area) and for seawater. The contribution of alpha radioactivity from radium-226 and uranium isotopes ranges from 8 to greater than 51 percent, except for seawater (less than 1 percent). Alpha radioactivity generally exceeds beta radioactivity in the ground-water samples. Water samples from rivers generally contain less radioactivity, and beta radioactivity usually exceeds alpha radioactivity. Beta radioactivity in rivers may be greater than alpha radioactivity because of less input from alpha-emitting members of the uranium-238 and thorium-232 series and more atmospheric contributions of beta emitters such as carbon-14. Almost all of the cosmogenic radionuclides are beta emitters (National Council on Radiation Protection and Measurements, 1975, p. 27).

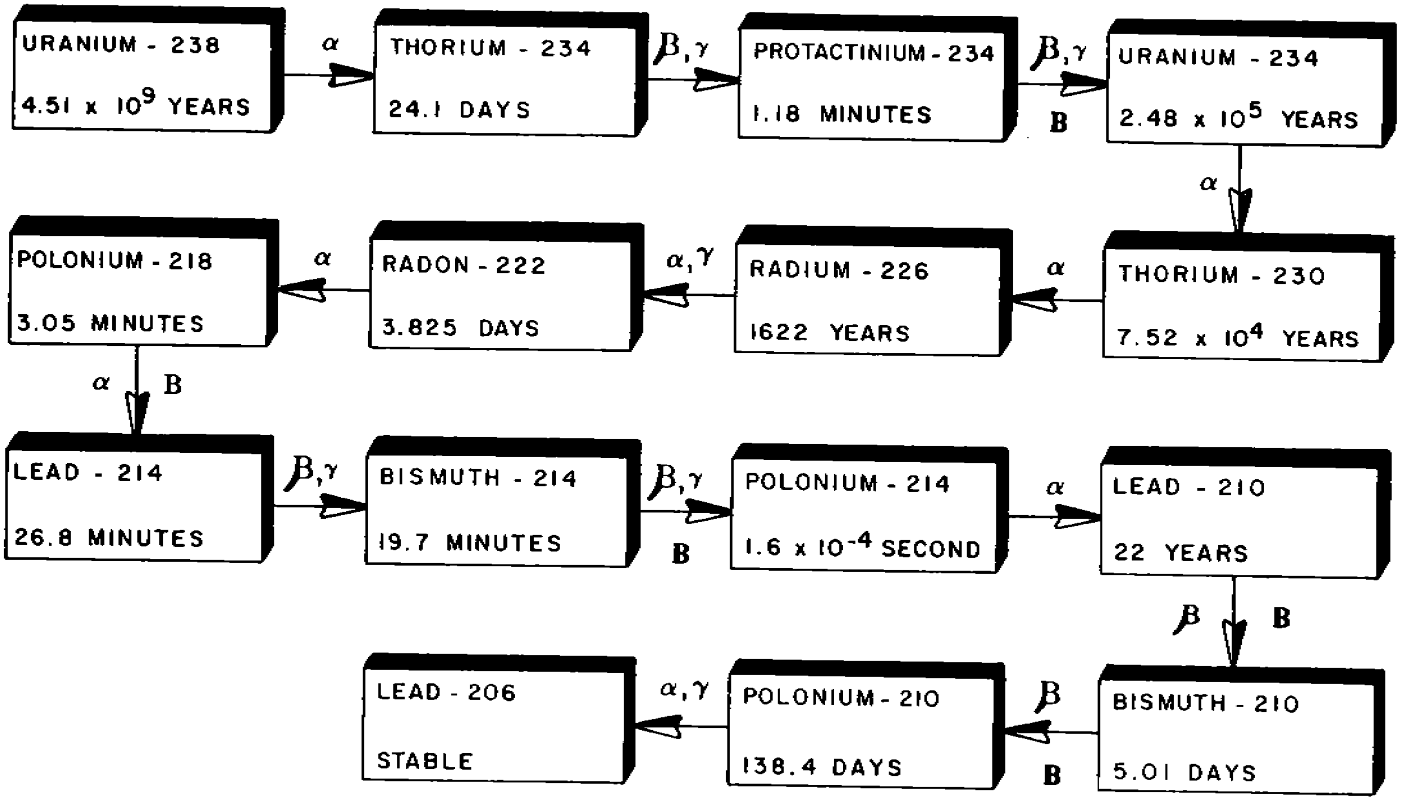


Figure 2.--Simplified diagram of predominant members of the uranium series. (B: minor branch that is not shown; α: alpha particle emission; β: beta particle emission; γ: gamma-ray emission.)

Table 3.--Selected radiochemical data for rivers and ground water in southwest Florida and for seawater

[pCi/L, picocuries per liter; GW, ground water; SW, surface water]

Site description	Site type	Dissolved alpha radio-activity (pCi/L as natural uranium)	Dissolved beta radio-activity (pCi/L as strontium-90)	Dissolved radium-226 (alpha radio-activity, in pCi/L)	Dissolved uranium (alpha radio-activity, in pCi/L)[1]	Percent of alpha acitvity from uranium and radium-226	Specific conductance (umho/cm at 25°C)
Sanibel water plant - raw water composite	GW	380	120	31	0.073	8	4,180
Rotunda west reverse osmosis plant - untreated	GW	180	63	28	0.04	16	12,400
City of Venice shallow wells, untreated	GW	48	14	7.8	0.05	16	1,222
Arcadia water supply, untreated	GW	42	17	8.2	0.007	20	655
City of Sarasota water supply, untreated	GW	21	9.5	3.5	0.093	17	1,042
McArthur EAM-1 near Sarasota	GW	26	9.9	*4.0	3.4	~28	728
Englewood water supply, untreated	GW	11	6.5	1.5	0.067	14	950
Peace River at Arcadia	SW	1.5	4.4	0.16	0.19	23	142
Hillsborough River near Zephyrhills	SW	<1.7	2.4	0.33	0.53	>51	300
Alafia River at Lithia	SW	<1.6	4.3	0.09	0.45	>34	248
Myakka River near Sarasota	SW	0.7	3.4	0.16	0.18	49	90
Gulf of Mexico at Siesta Beach	SW	<590	510	0.4	<4.0	<1	--
Average composition of seawater	SW	-	-	0.09[2]	3.33[3]	-	--
Gulf of Mexico	SW	-	-	0.1[3]	3.39[2]	-	--

[1] Assumes natural equilibrium abundances of uranium isotopes and equal radioactivities of uranium-238 and uranium-234.

[2] Joseph and others (1971).

[3] Osmond (1964).

* Planchet-count method, including all alpha-emitting radium isotopes.

Data in table 3 show that 28 percent or less of the alpha radioactivity in ground-water samples can be attributed to uranium isotopes and radium-226. Carbon-14, potassium-40, and rubidium-87 that are important sources of natural radioactivity are not alpha emitters. Therefore, radionuclides that contribute the bulk of the alpha radioactivity are probably alpha-emitting daughters of radon-222.

GEOLOGY AND DEPOSITION OF URANIUM MINERALS

Uranium and other members of the uranium series are commonly associated with a carbonate fluorapatite of marine origin. Uranium and other rare earths dissolved in seawater often replace the calcium in the carbonate fluorapatite, a complex phosphatic mineral of the phosphorite deposits of Florida. The phosphorite is notably devoid of separate uranium mineral phases (Altschuler and others, 1958).

The initial deposition of apatite was in a pellet precipitate or a replacement product in limestone or dolomite pebbles. This deposition was contemporaneous with the deposition of the Hawthorn Formation and to some extent the Tampa Limestone in shallow seas (table 4). Uranium substitution of calcium in the apatite also occurred at the same time. The sediments of the Hawthorn Formation were reworked numerous times by sea currents, concentrating the pellets and pebbles, accounting for the beds of uranium-bearing phosphatic pebbles and sand found in the Hawthorn Formation.

The Hawthorn Formation was exposed and extensively weathered during late Miocene time. This exposure developed an irregular topography and a surficial accumulation of primary phosphate pellets and pebbles from the Hawthorn Formation as well as secondary phosphatized limestone pebbles. The stratigraphic unconformity between the underlying Hawthorn Formation and the overlying Tamiami Formation, and in particular the Bone Valley Formation, indicates a record of later marine transgression with repeated pulses of reworking. This further concentrated the phosphatic pellets and pebbles from the Hawthorn Formation residue. Exposure to seawater probably allowed additional uptake of uranium.

The Bone Valley Formation, the primary source of phosphorite, later underwent extensive lateritic weathering that concentrated phosphate and uranium in the sand and pebble conglomerate of the basal part of the Bone Valley Formation.

Kaufman and Bliss (1977) stated that economically mineable phosphate deposits are present only in northeastern Sarasota County. Phosphatic deposits elsewhere in the county are thin or missing, or where thick, are confined to small isolated areas. The lack of mineable deposits of phosphate in the Bone Valley Formation in much of Sarasota County is an indication of less phosphate and therefore less uranium series rare earths in the surficial aquifer.

Uranium-rich zones and therefore radium-226-rich zones should be present in the more clastic (sand and gravel) zones of the Hawthorn, Tamiami, and Bone Valley Formations wherever phosphatic material is concentrated. The phosphatic pellets and pebbles in the Hawthorn Formation probably were concentrated by long shore currents, whereas those in the Tamiami and Bone Valley Formations were concentrated in strand lines as beach deposits. Both methods of concentration would produce linear deposits of uranium-bearing phosphatic material. Thus, one could expect uranium and accompanying radioactivity in the ground water to also exhibit a linear pattern with zones of low radioactivity between zones of high radioactivity within the same water-bearing unit.

Table 4.--Hydrogeologic units penetrated by water wells in Sarasota County

[Modified from Sutcliffe and Thompson, 1983]

Hydrogeologic unit	Zone	Formation	Characterist.	Thickness (feet)	Use	Well yields
Surficial aquifer	Surficial aquifer	Surface sand and terrace deposits.	Sand and sandy clay, fossiliferous, alluvial deposit.	10-80	Widely used in the Venice-Englewood area for domestic and public supplies; elsewhere usually cased off.	Up to 100 gal/min.
		Caloosahatchee Marl	Limestone, marl, sand, and gravel or quartz and phosphate, fossiliferous, including bone fragments.	0-10	Widely used in the Venice-Englewood area for domestic and public supplies; elsewhere cased off or absent.	Up to 100 gal/min.
		Bone Valley Formation	Sand and gravel, quartz and phosphate, clayey, fossiliferous, including bone fragments.	0-50	Widely used in the Venice-Englewood area for domestic and public supplies; elsewhere cased off or absent.	Up to 100 gal/min.
Intermediate aquifer and confining beds					Confining layer.	
	1	Tamiami Formation	Clay, green, sandy; clay, green and gray; limestone, tan; sandstone, gray; phosphate pebbles abundant near base. Local dark-gray waxy clay ("Venice Clay") at base.	0-120	Widely used in the Venice-Englewood area for domestic and public supplies; elsewhere usually cased off or absent.	Up to 150 gal/min.
					Confining layer.	
	2	Hawthorn Formation	Clay, limestone, and dolomite stringers, gray, green, and blue, phosphatic; interbedded with fossiliferous, sandy, limestone, silt, and sand.	200-400	Highly developed in coastal area for domestic, public supply, and home irrigation.	Upper part, 25-30 gal/min. Lower part, up to 500 gal/min.
					Confining layer.	
	3				Widely used for irrigation.	Average 250 gal/min. Up to 500 gal/min.
		Tampa Limestone	Limestone, white, gray, and tan; sandy, generally hard, dense, phosphatic, partly silicified, fossiliferous, solution cavities; clay, gray.	100-250	Widely used for irrigation.	Average 250 gal/min. Up to 500 gal/min.
Floridan aquifer system					Confining layer.	
	4	Suwannee Limestone	Limestone, light-colored, granular, soft to hard, porous, crystalline, dolomitic, very fossiliferous.	120-420	Widely used for irrigation.	500-1,500 gal/min.
		Ocala Limestone	Limestone, light-colored, granular, soft, chalky, interbedded with porous, hard, dense, fossiliferous limestone and thick, brown, crystalline, dolomitic limestone.	300-400	Semiconfining layer.	Yields little water.
	5	Avon Park Limestone	Limestone, light-colored, tan and brown, granular, soft and hard, very porous, interbedded with fossiliferous, crystalline limestone and dolomitic limestone.	600-700	Used only where large yields needed, mostly for irrigation.	1,500-2,000 gal/min.
Lower semiconfining unit		Lake City Limestone	Dolomite, limestone, gypsum, and anhydrite.		No use.	No use.

HYDROGEOLOGIC FRAMEWORK

Sarasota County is underlain by a thick sequence of sedimentary rocks whose lithology and structure control the occurrence, movement, and quality of ground water. The principal hydrogeologic units are (1) the surficial aquifer, (2) the intermediate aquifer and confining beds, (3) the Floridan aquifer system, and (4) the lower confining bed (table 4). In much of Sarasota County, the Floridan aquifer system, the major source of ground water in most of west-central Florida, does not contain freshwater; therefore, most freshwater in the county is supplied from wells that tap the surficial and intermediate aquifers. The Floridan aquifer system, however, does provide water for irrigation in the interior areas.

Sutcliffe and Thompson (1983) divided the intermediate aquifer and confining beds and the Floridan aquifer system into hydrologic zones that identify the productive water-bearing intervals. The hydrologic zone designations (table 4) of Sutcliffe and Thompson (1983) were used to identify the water-bearing intervals (in addition to the surficial aquifer) that were sampled in phase one of this study. Zone 1 consists of the Tamiami Formation and is separated from the underlying zone 2 by the "Venice clay." Zone 2 is the upper part of the Hawthorn Formation. Zone 2 is separated from zone 3 by green, gray, and blue interbedded clays near the middle part of the Hawthorn Formation. Zone 3 consists of the lower part of the Hawthorn Formation and the upper part of the Tampa Limestone. Gray clay beds that make up the middle and lower sections of the Tampa Limestone act as the confining bed between zones 3 and 4. Zone 4 consists of the Suwannee Limestone and the upper part of the Ocala Limestone. In the Sarasota area, the Ocala Limestone is a chalky, rather dense, and fossiliferous limestone. It has very little porosity in its middle and lower parts and acts as a confining bed for zone 5, the Avon Park Limestone. Zones 4 and 5 constitute the Floridan aquifer system in this area. More detailed information on the hydrogeology of this area has been published in a report by Wolansky (1983).

Surficial Aquifer

The surficial deposits consist of sands and terrace deposits, the Caloosahatchee Marl, and the Bone Valley Formation. These deposits are discontinuous, forming a surficial aquifer of variable thickness and permeability. Ground water generally occurs in deposits of sand, gravel, shell, phosphate, and limestone and is unconfined.

Characteristics of the surficial aquifer vary greatly. In the northeastern part of the county where the Bone Valley Formation is present, the aquifer consists mostly of medium- to fine-grained, well-sorted, quartz sand and phosphate gravel and ranges in thickness from about 10 to 60 feet. In the western and southern parts, the aquifer consists of sand, sandy limestone, marl, and shell fragments and ranges in thickness from about 1 to 20 feet. The permeable parts of the aquifer grade downward into a sandy silt, clay, or marl that acts as a confining bed for the underlying artesian zones.

Water levels in the surficial aquifer range from near land surface in coastal and low-lying areas to 10 feet or more below land surface in higher areas. In flat, poorly drained areas, average depth to water is less than 3 feet. Seasonal fluctuations of water levels are generally less than 5 feet. Water levels are generally lowest in May or June and highest in September or October.

The direction of ground-water flow is generally west and south. The configuration of the water table in the surficial aquifer is similar to that of land surface and locally is controlled by surface drainage. The water table is at or near sea level along the coast and increases to altitudes of about 90 feet above sea level in the northeastern part of the county.

The surficial aquifer is used as a source of water near the coast and in the southern parts of the county where water from the deeper aquifers contains more than 500 mg/L of dissolved solids. Elsewhere, the surficial aquifer generally is not used as a source of water. Water from the surficial aquifer is used primarily for domestic supplies, lawn and garden irrigation, and watering stock; however, several public supplies in the southern part of the county obtain some water from the surficial aquifer.

Most wells that penetrate the surficial aquifer are small-diameter, drive-point wells that yield from 5 to 20 gal/min. A few wells, 3 to 4 inches in diameter, are completed with open end and yield as much as 50 gal/min. Specific capacity ranges from 1 to 60 (gal/min)/ft and averages about 10 (gal/min)/ft (Wolansky, 1983). Public-supply wells in the Englewood area are 4 to 6 inches in diameter and have 20 feet of screen open to the permeable sand. The wells yield up to 60 gal/min (Sutcliffe and Thompson, 1983).

Water from the surficial aquifer generally contains less than 500 mg/L of dissolved solids except in areas adjacent to the coast, along tidally affected streams and canals, and in the vicinity of the Myakka River estuary (fig. 3). Concentrations of sulfate and chloride greater than 250 mg/L occur principally in coastal and southern areas (Wolansky, 1983).

Intermediate Aquifer and Confining Beds

The intermediate aquifer in Sarasota County consists of several water-bearing zones and confining beds in the Tamiami and Hawthorn Formations and in the Tampa Limestone (table 4). The total thickness of the intermediate aquifer water-bearing zones and confining beds ranges from about 300 feet in the northern part of the county to more than 400 feet in the southern part.

Water-bearing zones (three in number) consist of discontinuous beds of permeable sand, gravel, shell, and limestone and dolomite in the Tamiami Formation, the upper part of the Hawthorn Formation, and the lower part of the Hawthorn Formation and the upper part of the Tampa Limestone. Confining beds consisting of sandy clay, clay, and marl are at the base of the Tamiami Formation, separate the upper and lower parts of the Hawthorn Formation, and are at the base of the Tampa Limestone.

The composite potentiometric surface of the water-bearing zones of the intermediate aquifer ranges from about 30 feet above sea level in northeastern Sarasota County to less than 10 feet above sea level in the coastal areas. Seasonal water-level fluctuations range from about 5 feet in the northeastern and coastal areas to about 20 feet near the Verna well field in northern Sarasota County.

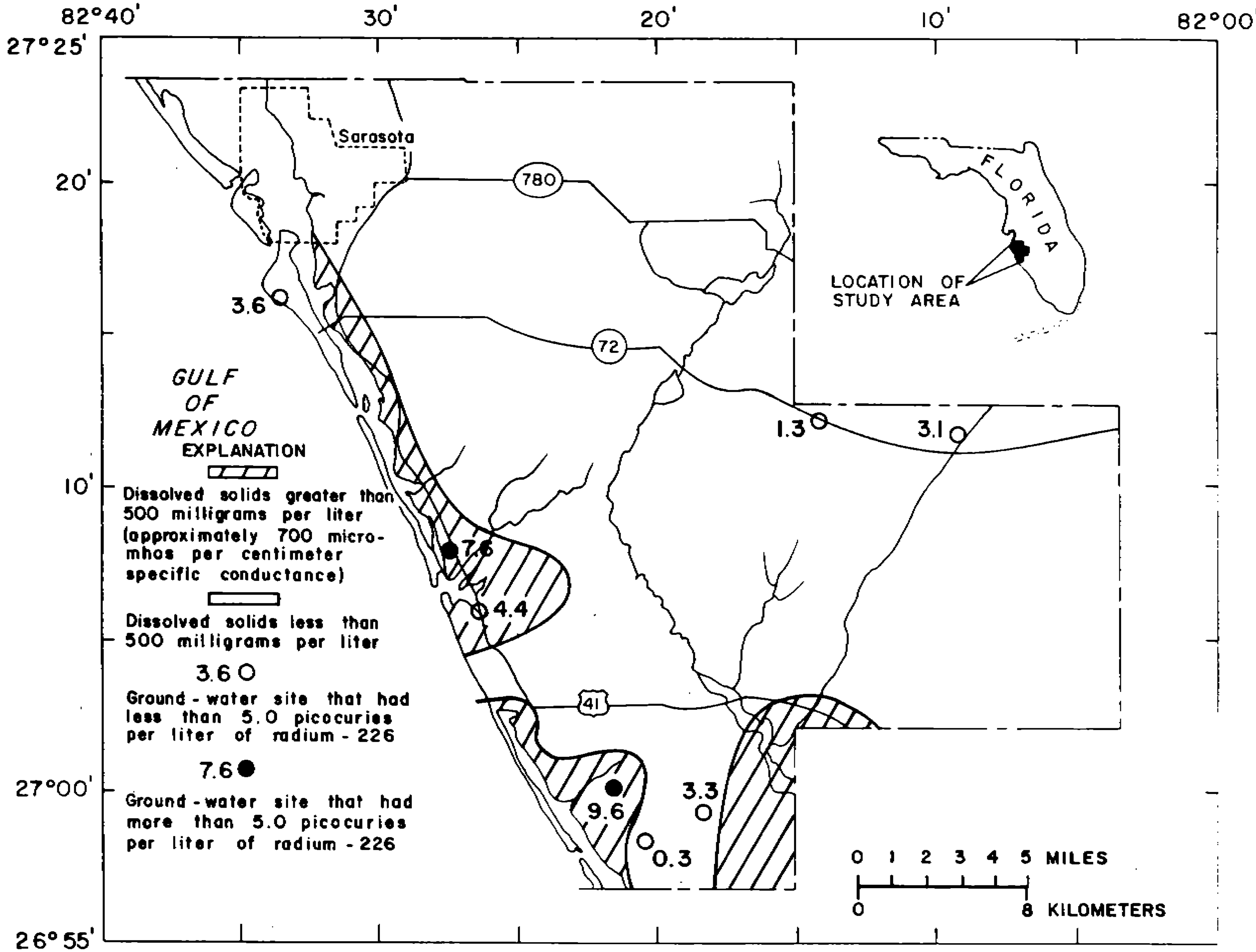


Figure 3.--Radium-226 radioactivities in water from the surficial aquifer and estimated areas where water in the surficial aquifer contains more than 500 milligrams per liter of dissolved solids. (Limits were estimated using data from Sutcliffe and Thompson, 1983; Wolansky, 1983; and Sutcliffe and Miller, 1981.)

Because dissolved solids in water of the underlying Floridan aquifer system are greater than in the intermediate aquifer, the intermediate aquifer is the most highly developed aquifer in the county and is used for domestic supply, irrigation, and public water supplies. The aquifer supplies most of the water for the cities of Sarasota and Venice, as well as other public water systems.

Zone 1

Most water in zone 1 in Sarasota County occurs in limestone, sandstone lenses with phosphate gravel, and shell beds that occur in the Tamiami Formation. The zone is thickest, as much as 120 feet, in the Venice-Englewood area (Sutcliffe and Thompson, 1983). The "Venice clay" that forms the confining bed in the lower part of the Tamiami Formation consists of 10 to 20 feet of waxy, dark green to gray clay or impure dolomite. The clay is an effective confining bed. Zone 1 extends north to the city of Sarasota and inland as much as 10 to 12 miles from the coastline (fig. 4). Wells completed in zone 1 are generally 3 to 4 inches in diameter and are finished as open hole in the limestone.

Chemically, the water is suitable for domestic and public supply except for an area along the coast north of Venice and in the area south of Englewood. It is widely used for domestic and public supplies in the Venice area, but elsewhere is usually cased off in the well or is absent.

Water levels in zone 1 are generally within 10 feet of land surface. Some wells developed in highly porous shell beds produce as much as 150 gal/min.

Zone 2

Zone 2 produces water from the upper part of the Hawthorn Formation. The zone consists of interbedded clay and limestone, sandstone, sand, silt, and dolomite. Fine-to-medium phosphate pebbles and grains occur throughout the zone. The beds occur at depths that range from 75 to about 200 feet below land surface. Zone 2 is highly developed for domestic lawn irrigation and public water supply. Water from the zone supplies about 60 percent of the potable water used in the populous coastal areas. Most wells drilled into zone 2 produce more than 25 gal/min. Larger diameter wells, 6 inches or more in diameter, produce more than 200 gal/min in most areas. Specific capacity ranges from 2 to 15 (gal/min)/ft and averages about 6 (gal/min)/ft (Wolansky, 1983). Areas where water from zone 2 contains less then 500 mg/L of dissolved solids are shown in figure 5.

Along the coast and in the southern part of the county, the water contains high concentrations of chloride and sulfate. Dissolved solids in these areas will exceed 500 mg/L (fig. 5).

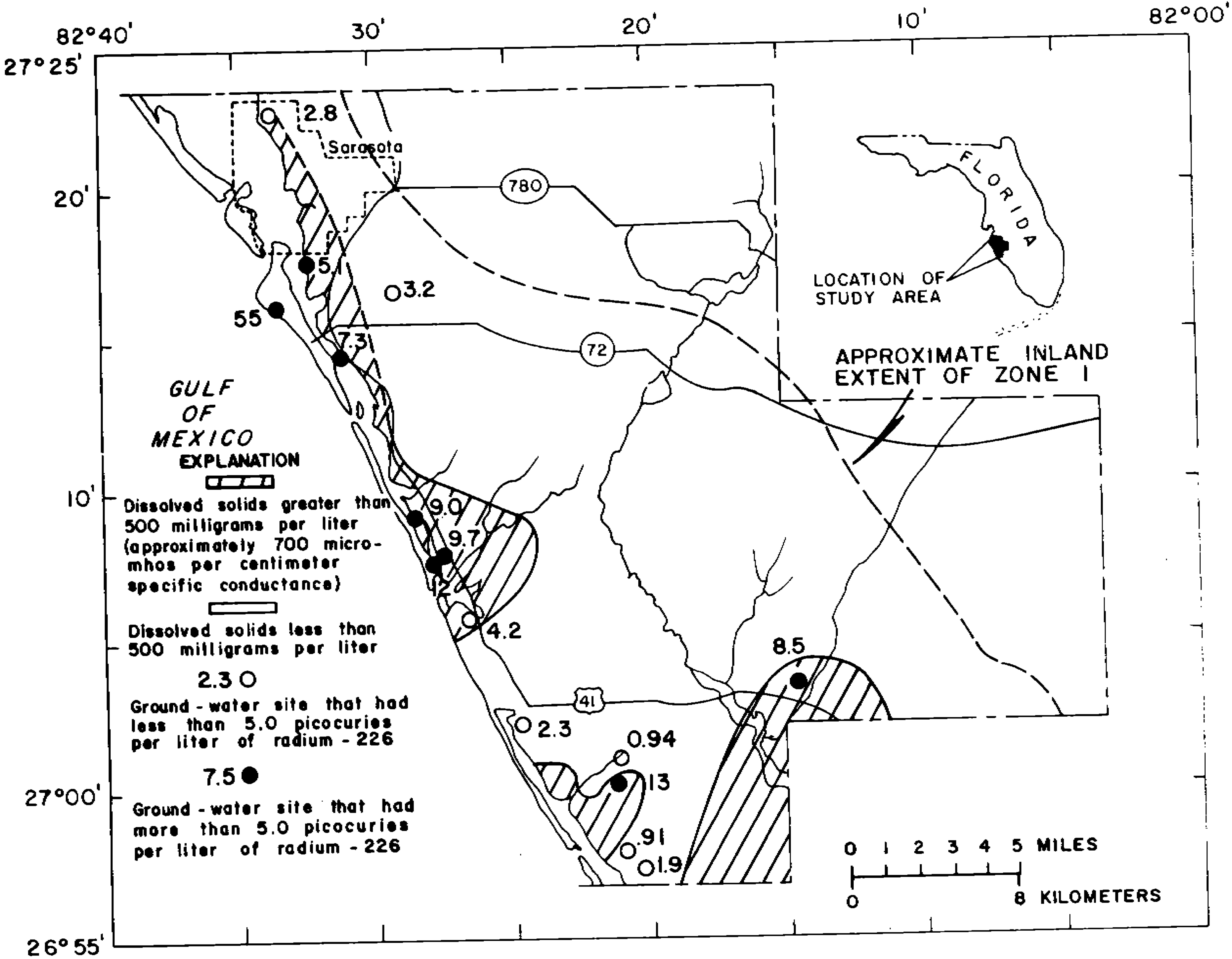


Figure 4.--Radium-226 radioactivities in water from zone 1 and estimated areas where water in zone 1 contains more than 500 milligrams per liter of dissolved solids. (Limits were estimated using data from Sutcliffe and Thompson, 1983; Wolansky, 1983; and Sutcliffe and Miller, 1981.)

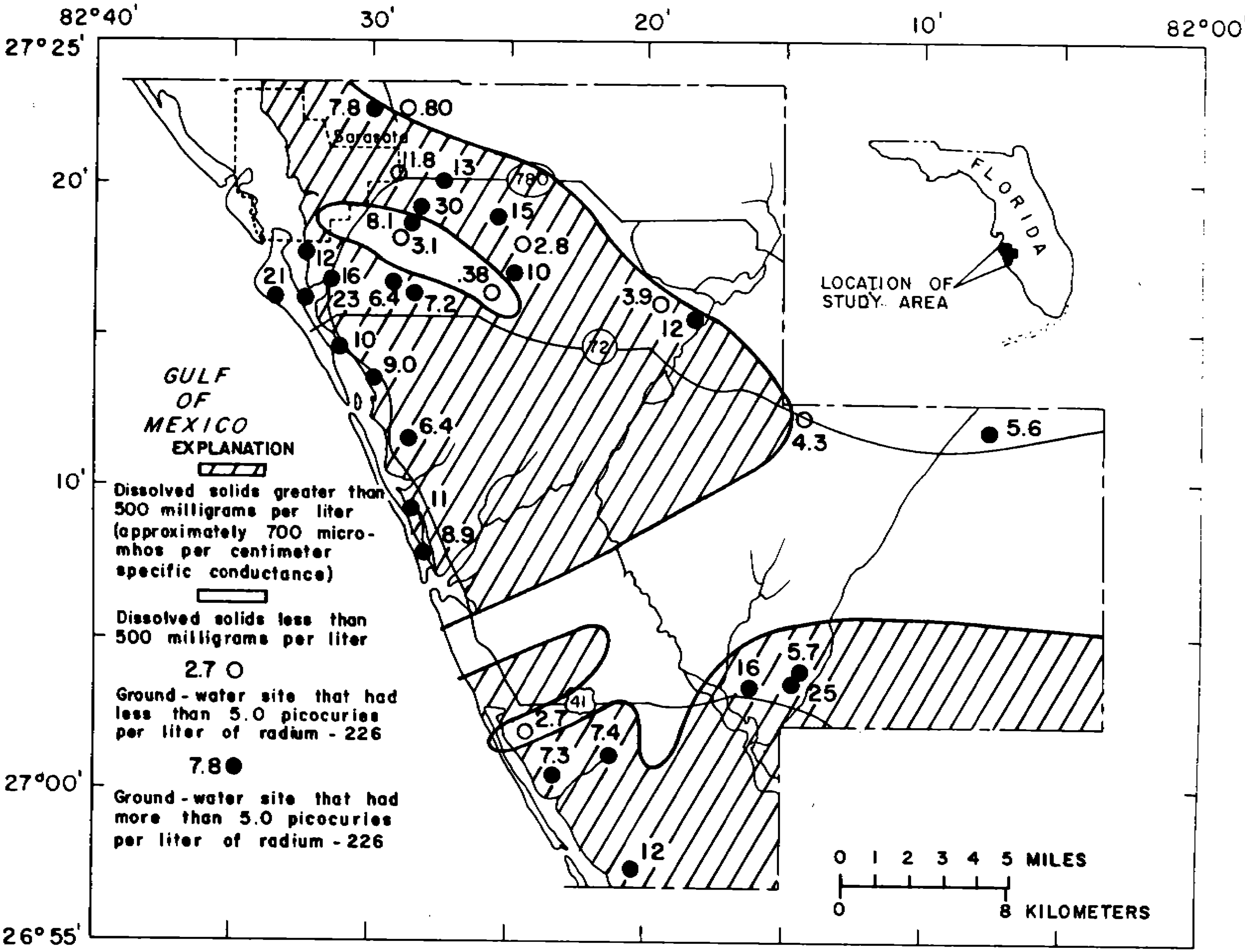


Figure 5.--Radium-226 radioactivities in water from zone 2 and estimated areas where water in zone 2 contains more than 500 milligrams per liter of dissolved solids. (Limits were estimated using data from Sutcliffe and Thompson, 1983; Wolansky, 1983; and Sutcliffe and Miller, 1981.)

Zone 3

Zone 3 consists of interbedded sand, clay, silt, limestone, and shell beds with occasional dolomite stringers in the lower part of the Hawthorn Formation and all but the basal part of the Tampa Limestone. The upper part of the Tampa Limestone grades downward into a gray, sandy, phosphatic marl or claystone. The lower part of the Tampa Limestone forms the confining bed between the intermediate aquifer and the Floridan aquifer system. Zone 3 occurs throughout Sarasota County, and the top of the zone ranges in depth from 150 to 300 feet below land surface. Wells open to zone 3 yield as much as 500 gal/min. Specific capacity ranges from 2 to 20 (gal/min)/ft and averages 10 (gal/min)/ft (Wolansky, 1983). Many large diameter irrigation wells in the eastern part of the county that are open to the underlying Floridan aquifer system are also open to zone 3. Zone 3 probably contributes about 20 percent of the yield of these wells. Water from zone 3 generally contains less than 500 mg/L of dissolved solids in the northeastern half of the county (fig. 6).

Floridan Aquifer System--Zones 4 and 5

The Floridan aquifer system in Sarasota County is composed of a thick, stratified sequence of limestone and dolomite that may include the Suwannee Limestone, Ocala Limestone, and the Avon Park Limestone. The top of the Floridan aquifer system generally coincides with the top of the Suwannee Limestone. Underlying the Floridan aquifer system is the lower confining bed that generally occurs in the Lake City Limestone where it contains evaporites (table 4).

The limestone and dolomite sequence generally functions as a single hydrogeologic unit; however, two distinct water-bearing zones are known to exist in the sequence. They are zone 4 (the Suwannee Limestone and the upper part of the Ocala Limestone) and zone 5 (the Avon Park Limestone). The highest permeability generally occurs at or near formation contacts. The zones are separated by beds of lower permeability that act as semiconfining beds that retard vertical movement of water within the aquifer.

The altitude of the top of the Floridan aquifer system ranges from about 400 feet below land surface in the northern part of the county to about 550 feet below land surface in the southern part. Average thickness of the Floridan aquifer system is about 1,400 feet.

The altitude of the average potentiometric surface of the Floridan aquifer system ranges from about 40 feet above sea level in southeastern Sarasota County to about 20 feet above sea level in northern Sarasota County. The regional gradient and direction of flow is west and northwest. The northwesterly flow is due to a depression in the potentiometric surface in Manatee County due to heavy pumping for irrigation.

The Floridan aquifer system yields less than 500 gal/min from wells smaller than 6 inches in diameter that partially penetrate the aquifer to about 5,000 gal/min from wells 12 inches or more in diameter that fully penetrate the aquifer. Most old wells are cased only to the first competent rock and are finished as open hole. Presently, new wells must be cased to greater depths depending on their purpose. Water in zones 4 and 5 of the aquifer contains less than 500 mg/L of dissolved solids in only a small part of eastern Sarasota County (fig. 7).

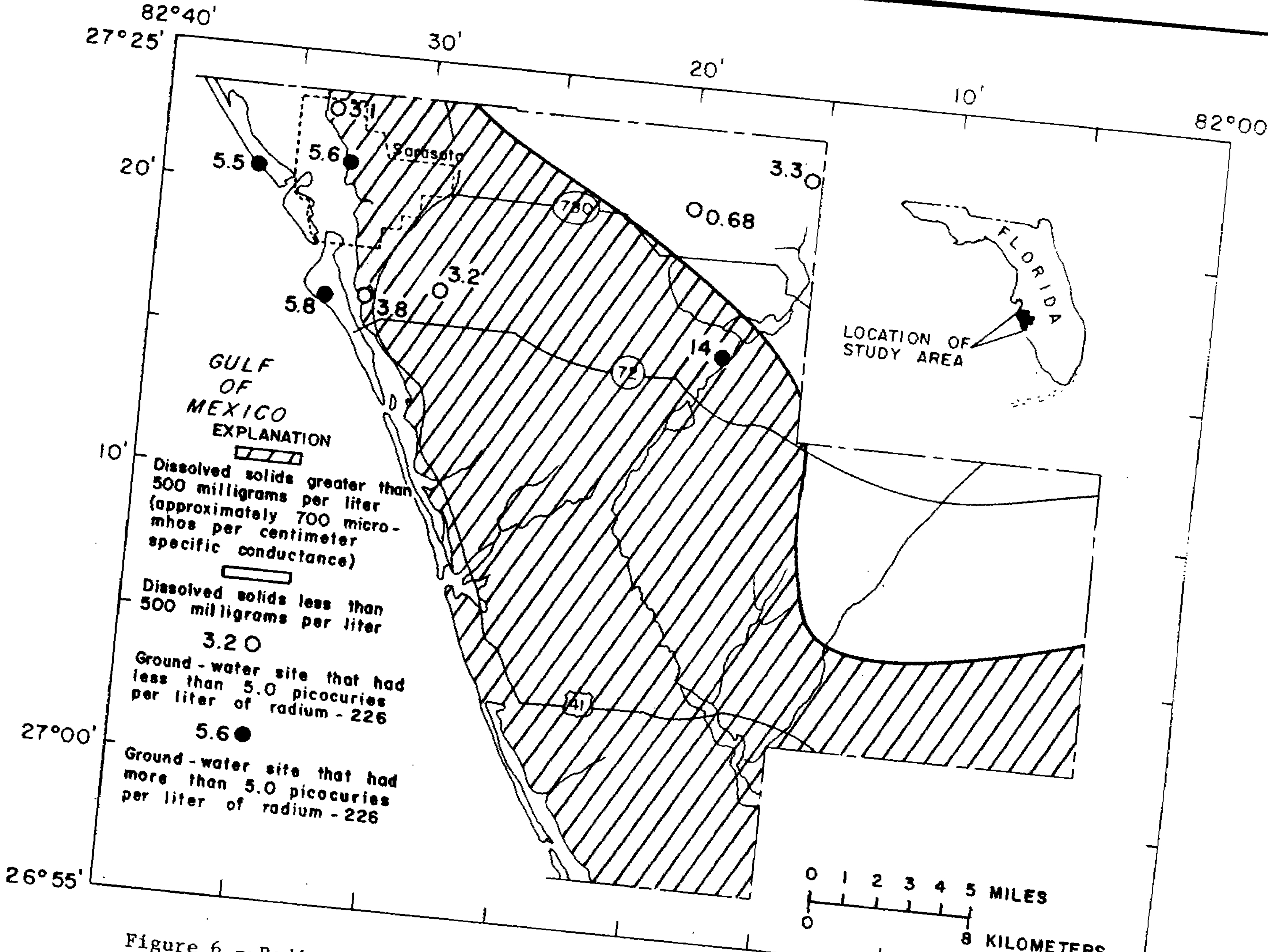


Figure 6.--Radium-226 radioactivities in water from zone 3 and estimated areas where water in zone 3 contains more than 500 milligrams per liter of dissolved solids. (Limits were estimated using data from Wolansky, 1983; and Sutcliffe and Miller, 1981.)

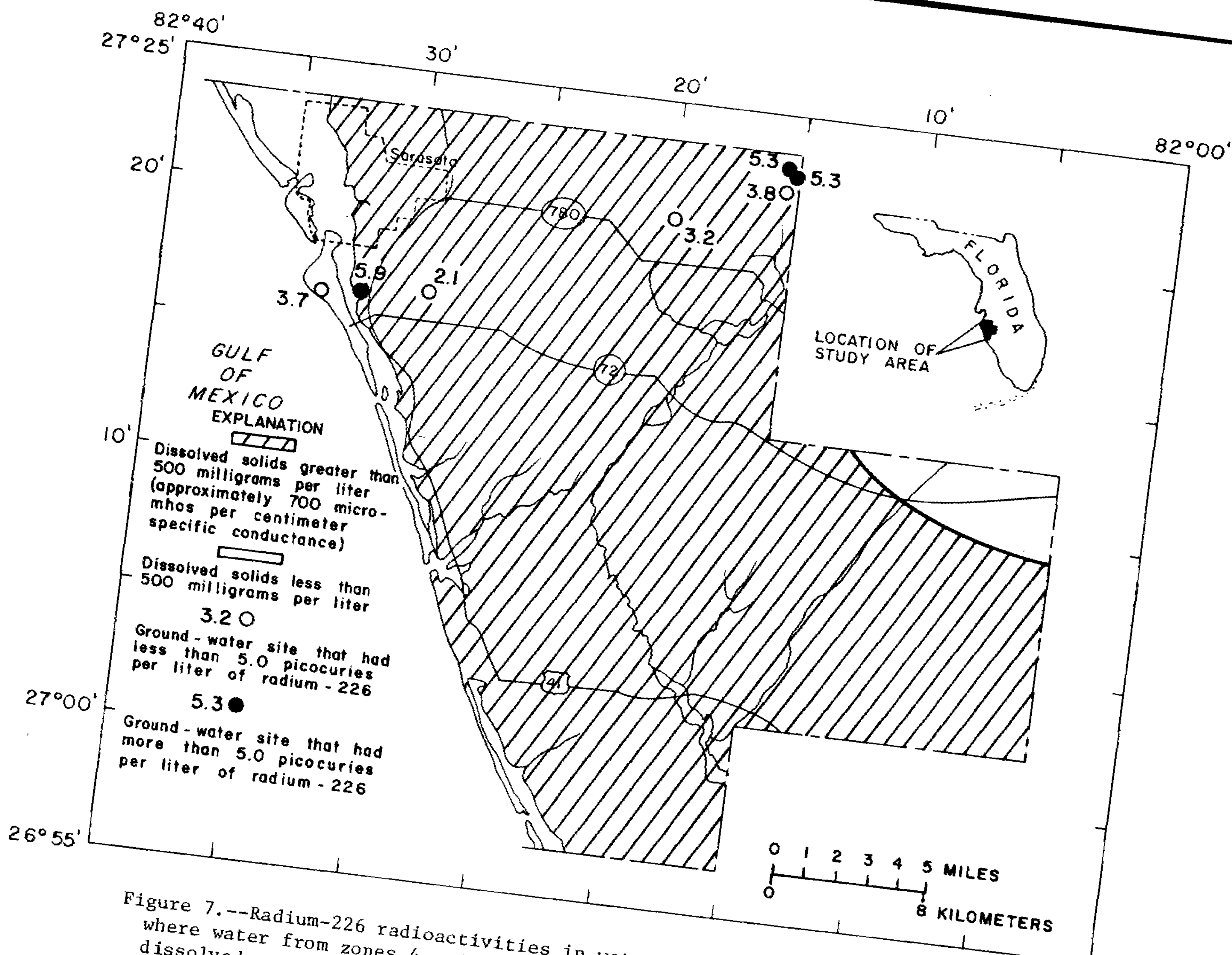


Figure 7.--Radium-226 radioactivities in water from zone 4 and estimated areas where water from zones 4 and 5 contains more than 500 milligrams per liter of dissolved solids. (Limits were estimated using data from Wolansky, 1983; and Sutcliffe and Miller, 1981.)

DISTRIBUTION OF RADIUM-226 IN GROUND WATER

The areal and vertical distributions of radium-226 radioactivity in waters of the aquifers underlying Sarasota County were examined to identify the water-bearing zones and, if possible, the areal extent of those zones that contain water of high radium-226 radioactivities. Distributions of radium-226 were also used in this report to interpret geochemical data and determine sources of radium-226 in water.

Areal Distribution of Radium-226

Radium-226 data were examined to define areas where radium-226 radioactivities in ground water were less than the 5.0-pCi/L limit for radium-226 plus radium-228. If radium-228 radioactivities are relatively minor compared to radium-226 radioactivities, such areas could be tapped for public supplies without treatment for removal of radium-226.

Figures 3 through 7 show radium-226 radioactivity for each well that sampled a single water-bearing zone. Wells shown in the figures were either open to only one zone or were sampled at several depths during drilling. If wells were sampled at more than one depth in the same zone, the average value for the zone was used in the figures. The distribution of data is heavily influenced by the location of most wells near coastal areas where much of the county's population is concentrated. Zone 2 is the common water-producing zone; consequently, more data were available for zone 2 than for other zones. Of 110 samples from 53 wells used to prepare the figures, 48 samples were from 33 wells in zone 2.

No definite patterns were discernible in the distribution of radioactivity in the water of the individual zones (figs. 3-7). When all zones are taken collectively, however, water containing more than 5.0 pCi/L appears to be more often associated with the more mineralized water of the coastal areas.

Vertical Distribution

The vertical distribution of radium-226 was evaluated to determine if any water-bearing zones existed that consistently contained water with less than 5.0 pCi/L of radium-226. If such zones exist, wells could be constructed to tap only zones with low radium-226 radioactivities. An understanding of vertical distributions of radium-226 also aids in interpretation of geochemical data and determination of sources of radium-226.

The Mann-Whitney nonparametric test (Snedecor and Cochran, 1967, p. 130) was used to test for differences at the 95-percent confidence level between populations of radium-226 data by individual water-bearing zones that are in contact with each other and by groups of zones. Test results between individual zones indicated that no significant differences existed in radium-226 radioactivity between the surficial aquifer and zone 1, between zones 1 and 2, or between zones 3 and 4. However, a significant difference was indicated between zones 2 and 3 (table 5). Uranium series radionuclides are present in phosphate nodules in the predominantly clastic materials above zone 3. Uranium-bearing apatite is less abundant and associated with limestone in zone 3 and is absent below zone 3. Test results of the combined data further indicated that zones 1 and 2 were significantly different from the combined data for zones 3 and 4 at the 95-percent confidence level using the Mann-Whitney test. This difference is due mainly to the 48 data for zone 2 that dominate the total of 67 data for zone 1 and zone 2. The only significant differences found between individual zones using the Mann-Whitney test occurred when data for zone 2 were compared with data for the surficial aquifer, zone 3, and zone 4. The results of these statistical tests should be used cautiously because the number of data and the spatial coverage within Sarasota County are limited, except for zone 2.

Table 5.--Statistics of central tendency for radium-226 radioactivities from discrete hydrologic zones

Zones	Radium-226, in pCi/L					Number of samples	Number of samples greater than 5.0 pCi/L	Number of wells
	Median	Geometric mean	Arithmetic mean	Minimum value	Maximum value			
Surficial aquifer	3.3	3.0	4.5	0.3	13	9	2	8
1	8.4	6.2	13.7	.91	110	19	11	15
2	8.8	7.8	10.5	.38	30	48	37	33
3	3.6	3.4	4.4	.16	14	22	7	9
4	3.8	3.4	3.7	1.3	5.9	12	4	7
1 and 2 combined	8.5	7.3	11.4	.38	110	67	48	41
3 and 4 combined	3.6	3.4	4.2	.16	14	34	11	12

Regression Analysis

Plots of radium-226 radioactivity versus specific conductance for each hydrologic zone are shown in figure 8. Radium-226 radioactivity is significantly correlated to specific conductance only in zones 1 and 2, zones that contain the highest concentrations of semiconsolidated pebble phosphate. For each 100-umho/cm increase in specific conductance in water from zones 1 and 2, there is a corresponding increase in radium-226 of 0.6 and 0.4 pCi/L, respectively. This increase compares favorably with results of Kaufmann and Bliss (1977, p. 69) who

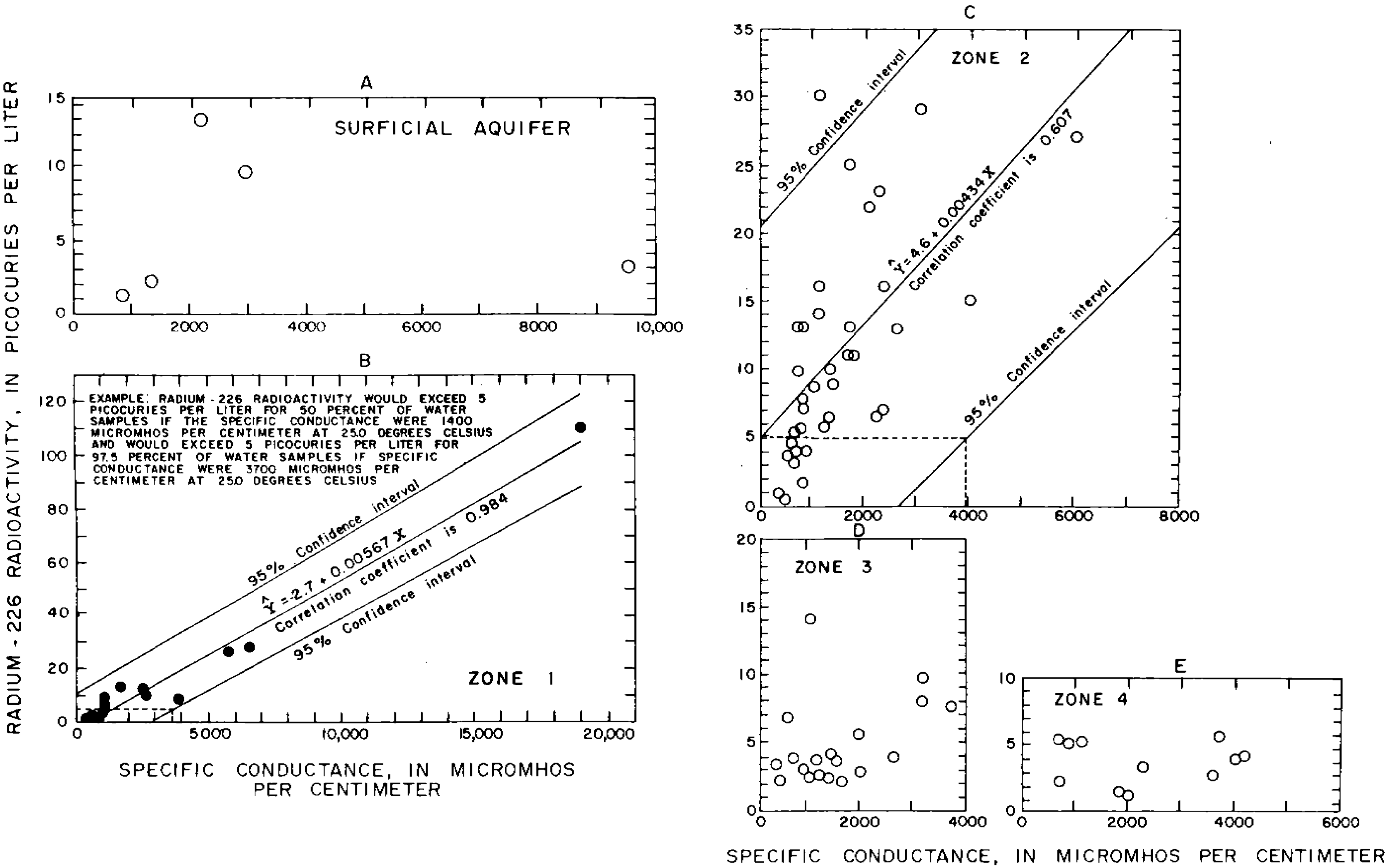


Figure 8.--Radium-226 radioactivity as a function of specific conductance in five water-bearing zones.

indicated that there was a 1-pCi/L increase in radium-226 in water from the Hawthorn Formation for each 100-mg/L increase in the concentration of dissolved solids. Based on data from Sutcliffe and Miller (1981), this equates to an 0.8-pCi/L increase in radium-226 for each 100-umho/cm increase in specific conductance.

Numbers of data pairs of radium-226 and specific conductance and correlation coefficients (R) are tabulated below for each hydrologic zone. Also given are probabilities of greater correlation coefficients (R) occurring by chance if the null hypothesis, rho = 0, holds. Rho represents the true correlation coefficient for a population rather than an estimator from a finite sample (R).

Zone	Number of data pairs	Correlation coefficient (R)	Probability of a greater R occurring by chance if rho = 0
Surficial	5	-0.07	0.91
1	17	.98	.00
2	36	.61	.00
3	19	.38	.11
4	11	.05	.89

Regression equations of the relation between radium-226 and specific conductance for water from zones 1 and 2 are also given in figure 8. The slopes of the regression lines for the relations in zones 1 and 2 are not significantly different at the 95-percent confidence level, but the elevations of the lines are significantly different; that is, one line is significantly higher than the other. Hence, data for these zones were not pooled to calculate a single regression line for both zones (Snedecor and Cochran, 1967). The regression lines provide the most likely radium-226 radioactivity for a given specific conductance. The lower confidence band gives the radium-226 radioactivity that will be exceeded for 97.5 percent of samples from each zone. Based on the regression analyses, radium-226 radioactivities in water from zone 1 would exceed 5.0 pCi/L for half of the water samples if the specific conductance is 1,400 umho/cm and would exceed 5.0 pCi/L for 97.5 percent of the samples with specific conductance of 3,700 umho/cm. Radium-226 radioactivities would exceed 5.0 pCi/L for half of the water samples from zone 2 if the specific conductance is 100 umho/cm and would exceed 5.0 pCi/L for 97.5 percent of samples with specific conductance of 3,900 umho/cm. Generally, in the surficial aquifer and in zones 1 and 2, water with a specific conductance of 700 umho/cm (about 500 mg/L dissolved solids) will contain less than 5.0 pCi/L of radium-226.

MECHANISMS CONTROLLING THE DISTRIBUTION OF RADIUM-226

As background for a discussion of three hypotheses that may describe the cause of elevated radium-226 activities in the study area, some relevant information is summarized as follows:

(1) Kaufmann and Bliss (1977) found that, in general, water in the surficial aquifer and Floridan aquifer system from areas with economically mineable phosphate deposits had lower concentrations of radium-226 than did water from Sarasota County.

(2) Only the northeast corner of Sarasota County has phosphate mineralization that is economically mineable (Kaufmann and Bliss, 1977).

(3) Factor analysis (see glossary) of data from all zones generated four factors from the data set of 25 complete analyses. Each factor is a combination of the original parameters, such as chloride and calcium concentrations, that emphasizes parameters that correlate with each other. Radium-226 was important in two factors. Factor one was comprised mainly of sulfate, calcium, magnesium, radium-226, bicarbonate, and strontium in decreasing order of importance. Factor three was comprised mainly of sodium, chloride, potassium, and radium-226 in decreasing order of importance. All correlations mentioned are direct except for bicarbonate ion that is inversely related.

(4) The greatest radium-226 radioactivities occur in zones 1 and 2. In these zones, phosphate commonly occurs as pebbles that are contained in semi-consolidated sands and clays. Radium-226 radioactivities in water from zones 1 and 2 are significantly correlated with specific conductance at the 95-percent confidence level.

(5) Radium-226 radioactivities tend to be lower in water from zones 3 and 4 than from zones 1 and 2. Only one radium-226 radioactivity out of 34 for zones 3 and 4 exceeded 10 pCi/L. Phosphate pebbles are less plentiful in zone 3 than in zones 1 and 2, and the pebbles are interbedded in limestone in zone 3. Phosphate pebbles are usually absent in zone 4.

(6) According to Zack (1980) increases in sodium and chloride inhibit the dissolution of fluorapatite.

(7) The occurrence of high radium-226 radioactivities in ground water in Sarasota County is a natural phenomenon that will probably not change rapidly with time except in areas where heavy pumping could cause increases in salinity and radium-226.

(8) High radium-226 radioactivities in zones 1 and 2 are associated with water that has high specific conductance. The same relation may also occur in the surficial aquifer and zone 3. Water that had high specific conductance was either a calcium magnesium strontium sulfate bicarbonate water or a marine-like water (sodium magnesium chloride). Steinkampf (1982) summarized the evolution of the calcium magnesium strontium sulfate bicarbonate type water found in Sarasota and surrounding counties. Dolomite $[CaMg(CO_3)_2]$ and gypsum $(CaSO_4 \cdot 2H_2O)$ dissolve and cause calcite $(CaCO_3)$ to precipitate, thus producing a water that is high in calcium, magnesium, and sulfate. Strontium is found where aragonite has recrystallized to calcite. Both types of high specific conductance water were associated with high radium-226 radioactivities.

With the aforementioned in mind, three hypotheses about the cause of high radium-226 radioactivities in Sarasota County and the relation between high radium-226 radioactivities and high specific conductance were evaluated. The first hypothesis is that gypsum, dolomite, and aragonite containing radium-226 dissolve and react to produce the calcium magnesium strontium sulfate bicarbonate waters of Sarasota County. Although some radium-226 will dissolve by this mechanism, it does not appear to be responsible for high radium-226 radioactivities. If this hypothesis were correct, an increase in sodium and chloride would not produce an increase in radium-226. What is observed is that increases in sodium and chloride are also correlated with an increase in radium-226 in water-bearing zones where phosphate ore is abundant.

A second hypothesis is that increases in ionic strength from marine-like waters or calcium magnesium strontium sulfate bicarbonate water lower activity coefficients and allow more phosphate ore and the radium-226 contained in the ore to dissolve. It is doubtful that this mechanism causes significant increases in radium-226. Increases in calcium concentrations from the calcium magnesium strontium sulfate bicarbonate water would tend to suppress the solubility of fluorapatite, which is a calcium mineral (common ion effect). The hypothesis that high ionic strengths of marine-like water lower activity coefficients and might allow more phosphate ore that hosts radium-226 to dissolve is controversial and could be questioned on the basis of Zack's (1980) work. According to Zack, hydroxylapatite and fluorapatite do not follow established classical principles of solubility-product equilibria, probably because of the formation of insoluble surface complexes, $Ca_2(HPO_4)(OH)_2$ and $Ca_2(HPO_4)F_2$. Zack's (1980) work suggests that increases in sodium chloride concentrations inhibit the dissolution of fluorapatite at least for short-term (4 days) experiments. L. N. Plummer (oral commun., 1983) and J. W. Ball (oral commun., 1983) believe that fluorapatite obeys solubility-product equilibria, although reliable data are difficult to acquire, and that the kinetics of dissolution and precipitation of fluorapatite are extremely slow.

A third hypothesis that may explain the high radium-226 radioactivities in ground water in Sarasota County involves alpha-particle recoil. A fraction of radium-226 could be ejected from a host mineral, especially in clay minerals and fine-grained phosphates, by alpha-particle recoil (4.68 million electron volts, MeV) as thorium-230 decays to radium-226 (T. F. Kraemer and D. F. Reid, written commun., 1983). If the ambient water is low in dissolved solids (minerals), the dissolved radium-226 is depleted by ion exchange or some other sorption mechanism. When high dissolved-solids concentration occurs in the ambient ground water, there is more competition with radium-226 for ion exchange or sorption sites, and more radium-226 accumulates in solution. Alpha-particle recoil would also help explain the high (up to 46,000 pCi/L) radon-222 radioactivities found in ground water by Kaufman and Bliss (1977) in Sarasota and nearby counties. Radon-222 is produced when radium-226 decays by alpha-particle emission (4.78 MeV). Because radon-222 is a noble gas that sorbs little and can not participate in ion exchange because it does not form ions, its concentration can increase to high levels in confined aquifers. Because radon-222 has a short half-life (3.8 days), an abundance of radium-226 in aquifer materials is required to maintain high radon-222 radioactivities.

T. F. Kraemer (oral commun., 1983) also stated that alpha recoil distances can reach several hundredths of a micrometer. This means that radium-226 could escape from clays associated with phosphate deposits and from the outer several hundredths of a micrometer of phosphate ore particles.

It seems likely that uranium in fluorapatite is the dominant source of radium-226 in the shallow zones because of higher concentrations of uranium in marine phosphorites (50 to 300 mg/kg) when compared to average concentrations of uranium in limestone in North America (2.2 mg/kg), silica (0.1 to 10 mg/kg), and feldspar (0.1 to 10 mg/kg) as reported by Wedepohl (1974). Monazite contains 500 to 3,000 mg/kg of uranium (Wedepohl, 1974) and may be an important source of radium-226 (and radium-228 because of its thorium-232 content) where black sands occur along the coast of Sarasota County between Venice and Englewood. Monazite contains 20 to 200 mg/kg of thorium (Wedepohl, 1974). All of these minerals are sources of radium-226 through alpha-particle recoil and dissolution in the zones in which they occur. The highest radium-226 radioactivities, however, are found in zones where fluorapatite is plentiful. Because of the low solubility of fluorapatite, alpha-particle emission is thought to be an important factor in causing high radium-226 radioactivities in Sarasota County.

Radium-226 radioactivities in deep zones may be the result of alpha-particle recoil and dissolution of host minerals such as calcite, gypsum, dolomite, and aragonite. Average concentrations of uranium in limestones and dolomites are about 2 mg/kg for samples from North America and Russia (Wedepohl, 1974). Ratios of uranium-234 to uranium-238 suggest that alpha-particle recoil is important in deep zones (Osmond and Cowart, 1977).

If dissolution of host minerals is the dominant mechanism for the dissolution of the members of the uranium series, the ratio of radioactivities of uranium-234 to uranium-238 will remain near 1.0 (Kraemer, 1981). This assumes that the uranium-234 to uranium-238 ratio is 1.00 in the host minerals. If alpha-particle recoil is important, then the ratio of radioactivities in solution will be greater than 1.0. Uranium-238 will not escape the host mineral by alpha-particle recoil because it is the parent radionuclide of the uranium series, and when it decays by alpha-particle emission, the recoiling nucleus is transformed from uranium-238 to thorium-234. Thorium-234 decays through protactinium-234 to uranium-234 by beta decay within a few months.

Osmond and Cowart (1977) determined ratios of uranium-234 to uranium-238 radioactivity on four water samples from wells less than 800 feet deep in Sarasota County. These ratios ranged from 1.39 to 1.77 and averaged 1.56. One well (24 feet deep) in reclaimed land at a phosphate mine in Polk County had a ratio of 2.4 (Miller and Sutcliffe, 1982). These ratios suggest that alpha-particle recoil is an important mechanism for introducing members of the uranium series into ground water in areas where phosphate ore exists.

Kaufmann and Bliss (1977) reported higher radium-226 radioactivities in Sarasota County than in the phosphate mining areas of Polk County. This could well be explained by the concept that, in Polk County, water of relatively low conductance comes in contact with the phosphate deposits and radium-226 is depleted from ground water by ion exchange or another sorption mechanism. Sarasota County contains more mineralized water that permits more radium-226 to remain in solution because there is more competition for ion exchange or sorption sites by higher concentrations of ions in solution.

Radium-226 is the first element in the uranium-238 decay series that is significantly soluble in a reducing environment that is low in sulfate. Kraemer and Reid (written commun., 1982) measured radium-226 radioactivities as high as 1,570 pCi/L in deep geothermal brines. Uranium is relatively insoluble under the reducing conditions found in most aquifers. High ratios of uranium to radium-226 radioactivity would be expected only where oxidizing conditions occur. In natural environments, oxidizing conditions are typically caused by the presence of oxygen gas. Where phosphate ore or other sources of the uranium series are abundant and mineralized water prevents radium-226 sorption, high radium-226 radioactivities generally occur.

EFFECTS OF WATER TREATMENT ON RADIUM-226 RADIOACTIVITIES

Dissolved radium-226 radioactivities are reduced by conventional water-treatment processes. Brinck and others (1978) found that the processes of reverse osmosis, cation exchange (using sodium zeolite), lime-soda-ash softening, and iron removal (precipitation and filtration, oxidation, or ion exchange) remove the major portion of radium-226 from water. The percent of radium-226 removed by these processes are given in table 6. Ground water in Sarasota County that is naturally high in radium-226 is also high enough in dissolved solids, chloride, or sulfate to exceed the National Secondary Drinking Water Regulations. Consequently, it is unlikely that public water supplies will produce water high in radium-226 because such water will normally be treated by at least one of the above mentioned processes, which will reduce radium-226 radioactivities to less than 5.0 pCi/L.

Reverse osmosis is used in Sarasota County to treat water for some small public supplies, such as schools and mobile home parks. Sorg and others (1980) found that, for eight reverse-osmosis plants in Sarasota County, 87 to 98 percent of the radium-226 radioactivity was removed. For the plants studied, the raw water had radium-226 radioactivities from 3.4 to 20.5 pCi/L. Radioactivities of the product water ranged from 0.14 to 2.01 pCi/L.

Reverse osmosis has been selected for use in many parts of Sarasota County because dissolved-solids concentrations are too high to meet the National Secondary Drinking Water Regulations (U.S. Environmental Protection Agency, 1977) of 500 mg/L. The reverse-osmosis process is an effective process for removal of both dissolved solids and radium-226. Most other conventional water-treatment processes do not significantly reduce dissolved-solids concentration.

Table 6.--Percent of radium-226 removal for conventional water-treatment processes

[Data from Brinck and others, 1978]

Treatment process	Percent radium-226 removal
Reverse osmosis	96
Sodium cation exchange	>90
Lime-soda-ash softening	75-96
Iron removal	11-53

POTENTIAL EFFECTS OF PHOSPHATE MINING

Phosphate mining rearranges the location of aquifer materials and alters the permeability of the reclaimed areas. One effect of phosphate mining may be exposure of residual phosphate ore in reclaimed areas and slime ponds to different pH, Eh, and ionic strength conditions. Such changes could increase the rate of accumulation of radium-226 and other radionuclides in ground water. As phosphate mining operations move closer to the west coast of Florida, there may be interest in using saline ground water or water from the Gulf of Mexico as process water. If the hypothesis that highly mineralized water results in high radium-226 radioactivities is correct, it might be prudent to avoid exposure of mining areas and reclaimed lands to highly mineralized water. Exposure to highly mineralized water can be avoided by using fresh waters for mining and transporting phosphate ores and slimes (clay waste with fine residual phosphate ore).

Phosphate mining in Sarasota County probably will not penetrate the aquifer system any deeper than zone 1. Consequently, major effects of mining would occur in the surficial aquifer and zone 1 (Tamiami Formation). Effects of mining probably would be indirect on hydrologic zones deeper than zone 1, such as those that result from heavy pumpage to meet increased water demands and dewatering of the surficial aquifer and zone 1 prior to mining. Dewatering may cause an upward leakage of water from zones 2 and 3 and the Floridan aquifer system. The effects of upward leakage on radium-226 radioactivities depends on the salinity and original radium-226 radioactivity of the water leaking upward.

SUMMARY AND CONCLUSIONS

1. Radium-226 radioactivity occurs in ground water throughout Sarasota County at levels in excess of the National Interim Primary Drinking Water Regulation of 5.0 pCi/L for the sum of radium-226 and radium-228.

2. The amount of radium-226 radioactivity varies with depth. The averages of radium-226 data for the water-bearing zones show that the greatest amounts occur in water from zone 2 (upper part of the Hawthorn Formation) that contained up to 30 pCi/L of radium-226 and from zone 1 (Tamiami Formation) that contained up to 110 pCi/L of radium-226. Zone 2 supplies about 60 percent of the water used in the populous coastal areas. The amount of radium-226 in water from the surficial aquifer and zone 3 (lower part of the Hawthorn Formation and Tampa Limestone) also can exceed the 5.0-pCi/L regulations for drinking water. All 12 water samples from zone 4 (Suwannee and upper Ocala Limestones) contained less than 6.0 pCi/L of radium-226.

3. The amount of radium-226 radioactivity in water samples from zones 1 and 2 is strongly correlated with specific conductance, and consequently, the dissolved-solids concentration in the water. Generally, in the surficial aquifer and in zones 1 and 2, water with a specific conductance of 700 umho/cm (about 500 mg/L dissolved solids) or less will contain less than 5.0 pCi/L of radium-226.

4. Water that contains radium-226 radioactivity in excess of limits set in the National Interim Primary Drinking Water Regulations usually was too mineralized to meet National Secondary Drinking Water Regulations for chloride, 250 mg/L; sulfate, 250 mg/L; or dissolved solids, 500 mg/L (approximately 700 umho/cm specific conductance). Most water containing high radium-226 radioactivity would be treated by reverse osmosis to reduce the dissolved-solids concentrations or by ion exchange water softening to reduce the hardness before public consumption. Both reverse osmosis and ion exchange reduce radium-226 below 5.0 pCi/L. Home water softeners use ion exchange and also will reduce radium-226 radioactivities.

5. High radium-226 radioactivities in water may occur where marine-like water or water that has high concentrations of magnesium, calcium, strontium, and sulfate is in contact with semiconsolidated deposits of phosphate pebbles in clastic materials. Semiconsolidated deposits of pebble phosphate occur in the surficial aquifer and in zones 1 and 2. In zone 3, the phosphate is contained in limestone and occurs in lesser amounts than in the zones above. In zone 4 and deeper zones, phosphate ore is absent.

6. Lower radium-226 radioactivities occur in ground water in the phosphate mining areas in Polk County than in Sarasota County. This may be explained by the hypothesis that water in contact with phosphate deposits in Polk County is lower in concentrations of dissolved ions and consequently can not compete with radium-226 for ion exchange and sorption sites as well as saline water in Sarasota County. Thus, in zones that contain phosphate ores, high dissolved solids seem to permit high radium-226 radioactivities.

7. Phosphate mining is a mechanical process that sorts and relocates aquifer materials but dissolves very little of this material. Exposure of residual clays associated with phosphate ore to mineralized water could result in release of radionuclides. Radium-226 dissolved by alpha-particle recoil could increase to higher concentrations in aquifers that contain mineralized water.

SELECTED REFERENCES

Altschuler, Z. S., Clarke, R. S., Jr., and Young, E. J., 1958, Geochemistry of uranium in apatite and phosphorite: Shorter contributions to general geology: U.S. Geological Survey Professional Paper 314-D, p. 45-90.

Bishop, E. W., 1960, Freshwater resources of Sarasota County, Florida: Board of Sarasota County Commission Research Report no. 1, 91 p.

Black, Crow and Eidsness, Inc., 1975, Wastewater treatment and disposal alternatives, City of Sarasota, January 20, 1975: Consulting Engineers Report for the city of Sarasota, 29 p.

Brinck, W. L., Schliekelman, D. L., Bennett, D. L., and others, 1978, Radium-removal efficiencies in water-treatment process: Journal of the American Water Works Association, v. 70, no. 1, p. 31-35.

Broecker, W. S., Li, Y. H., and Cromwell, J., 1967, Radium-226 and radon-222: Concentration in Atlantic and Pacific Oceans: Science, v. 158, no. 3806, p. 1307-1310.

Clark, W. E., 1964, Possibility of salt-water leakage from proposed intercoastal waterway near Venice, Florida, well field: Florida Geological Survey Report of Investigations 38, 33 p.

Eppert, H. C., 1966, Stratigraphy of the Upper Miocene deposits in Sarasota County, Florida: Tulane Studies in Geology, v. 4, no. 2, p. 49-61.

Fitzgerald, J. E., Jr., Guimond, R. J., and Shaw, R. A., 1976, A preliminary evaluation of the control of indoor radon daughter levels in new structures: Washington, D.C., U.S. Environmental Protection Agency, Office of Radiation Programs, Criteria and Standards Division Report no. EPA-520/4-76-018, 62 p.

Florida Department of Health and Rehabilitative Services, 1976, Study of radiation environment, Sarasota County, Florida: Unpublished report in files of the Sarasota County Health Department, dated May 20, 1976, 10 p.

Friedlander, G., Kennedy, J. W., and Miller, J. M., 1964, Nuclear and radiochemistry: New York, John Wiley and Sons, 585 p.

Guimond, R. J., and Windham, S. T., 1975, Radioactivity distribution in phosphate products, by-products, effluents, and wastes: Washington, D.C., U.S. Environmental Protection Agency, Office of Radiation Programs, Criteria and Standards Division Technical Note ORP/CDS-75-3, 30 p.

Hutchinson, C. B., 1975, Effects of strip mining on shallow aquifer systems in phosphate district: in Geological Survey Research 1975: U.S. Geological Survey Professional Paper 975, Chapter A, p. 89.

Irwin, G. A., and Hutchinson, C. B., 1976, Reconnaissance water sampling for radium-226 in central and northern Florida, December 1974-March 1976: U.S. Geological Survey Water-Resources Investigations 76-103, 16 p.

Joseph, A. B., Gustafson, P. F., Russell, I. R., Shuert, E. A., Volchok, H. L., and Tamplin, A., 1971, Sources of radioactivity and their characteristics: in National Academy of Sciences, Radioactivity in the marine environment: Chapter 2, p. 6-41.

Joyner, B. F., and Sutcliffe, H., Jr., 1976a, Water resources of the Myakka River basin area, Florida: U.S. Geological Survey Water-Resources Investigations 76-58, 87 p.

----- 1976b, Salt-water contamination in wells in the Sara-Sands area in Siesta Key, Sarasota County, Florida: Journal of the American Water Works Association, v. 59, no. 12, p. 1504-1512.

Kaufmann, R. F., and Bliss, J. D., 1977, Effects of phosphate mineralization in the phosphate industry on radium-226 in ground water in central Florida: Washington, D.C., U.S. Environmental Protection Agency Report no. EPA/520-6-77-010, 115 p.

Kraemer, T. F., 1981, ^{234}U and ^{238}U concentration in brine from geopressured aquifers of the northern Gulf of Mexico Basin: Earth and Planetary Science Letters, no. 56, p. 210-216.

Miller, R. L., and Sutcliffe, H., Jr., 1982, Water-quality and hydrogeologic data for three phosphate industry waste-disposal sites in central Florida, 1979-80: U.S. Geological Survey Water-Resources Investigations 81-84, 77 p.

National Council on Radiation Protection and Measurements, 1975, Natural background radiation in the United States: Washington, D.C., NCRP Report no. 45, 163 p.

----- 1976, Environmental radiation measurements: Washington, D.C., NCRP Report no. 50, 246 p.

Osmond, J. K., 1964, The distribution of the heavy radioelements in rocks and waters of Florida: in Adams, J. A. S., and Lowder, W. M., eds., The natural radiation environment: Chicago, University of Chicago Press, p. 153-159.

Osmond, J. K., and Cowart, J. B., 1977, Uranium series isotopic anomalies in thermal ground waters from southwest Florida: in Smith, D. L., and Griffin, G. M., eds., The geothermal nature of the Floridan Plateau: Florida Bureau of Geology Special Publication 21, p. 131-147.

Plummer, L. N., Jones, G. F., and Truesdell, A. H., 1978, WATEQF-a fortran IV version of WATEQ, a computer program for calculating chemical equilibrium of natural waters: U.S. Geological Survey Water-Resources Investigations 76-13, 63 p.

Samuals, L. D., 1964, A study of environmental exposure to radium in drinking water: in Adams, J. A. S., and Lowder, W. M., eds., The natural radiation environment: Chicago, University of Chicago Press, p. 239-251.

Snedecor, G. W., and Cochran, W. G., 1967, Statistical methods: Ames, Iowa, The Iowa State University Press, 6th ed., 593 p.

Sorg, T. J., Forbes, R. W., and Chambers, D. S., 1980, Removal of radium-226 from Sarasota County, Florida, drinking water by reverse osmosis: Journal of the American Water Works Association, v. 72, no. 4, p. 230-237.

Steinkampf, W. C., 1982, Origins and distribution of saline ground waters in coastal southwest Florida: U.S. Geological Survey Water-Resources Investigations 82-4052, 34 p.

Stringfield, V. T., 1933, Ground-water resources of Sarasota County, Florida, and exploration of artesian wells in Sarasota County, Florida: Florida Geological Survey 23rd-24th Annual Report.

Stumm, W., and Morgan, J. J., 1970, Aquatic chemistry - an introduction emphasizing chemical equilibria in natural waters: New York, Wiley Interscience, 583 p.

Sutcliffe, H., Jr., 1975, Appraisal of the water resources of Charlotte County, Florida: Florida Bureau of Geology Report of Investigations 78, 53 p.

Sutcliffe, H., Jr., and Joyner, B. F., 1966, Packer testing in water wells in Sarasota County, Florida: Ground water, v. 4, no. 2 p. 23-27.

----- 1968, Test well exploration in the Myakka River basin area, Florida: Florida Geological Survey Information Circular 56, 61 p.

Sutcliffe, H., Jr., and Miller, R. L., 1981, Data on ground-water quality with emphasis on radionuclides, Sarasota County, Florida: U.S. Geological Survey Open-File Report 80-1223, 13 p.

Sutcliffe, H., Jr., and Thompson, T. H., 1983, Occurrence and use of ground water in the Venice-Englewood area, Sarasota and Charlotte Counties, Florida: U.S. Geological Survey Open-File Report 82-700, 59 p.

Thatcher, L. L., Janzer, V. J., and Edwards, K. W., 1977, Methods for determination of radioactive substances in water and fluvial sediments: U.S. Geological Survey Techniques of Water-Resources Investigations, Book 5, Chapter A5, 95 p.

U.S. Atomic Energy Commission, 1967, Nuclear terms - a brief glossary: Oak Ridge, Tennessee, Division of Technical Information Extension, 80 p.

U.S. Environmental Protection Agency, 1975, Preliminary findings radon daughter levels in structures constructed on reclaimed Florida phosphate land: U.S. Environmental Protection Agency Technical Note QRP/CSD-75-4, 40 p.

----- 1976, National interim primary drinking water regulations: EPA-570/9-76-003, 150 p.

----- 1977, National secondary drinking water regulations: Federal Register, v. 42, no. 62, p. 17143-6.

Wahba, C. M., Hill, H., and More, A. I., 1980, Phosphoric acid-outline of the industry: London, The British Sulphur Corporation Limited, 78 p.

Weast, R. C., editor, 1976, Handbook of chemistry and physics: Cleveland, CRC Press, 57th ed., 2,391 p.

Wedepohl, K. H., editor, 1974, Handbook of geochemistry: New York, Springer-Verlag, p. 92-A-1f.

Wilson, W. E., 1977, Ground-water resources of De Soto and Hardee Counties, Florida: Florida Bureau of Geology Report of Investigations 83, 102 p.

Wolansky, R. M., 1983, Hydrogeology of the Sarasota-Port Charlotte area, Florida: U.S. Geological Survey Water-Resources Investigations 82-4089, 48 p.

Zack, A. L., 1980, Geochemistry of fluoride in the Black Creek aquifer system of Horry and Georgetown Counties, South Carolina--and its physiological implications: U.S. Geological Survey Water-Supply Paper 2067, 40 p.

*U.S. GOVERNMENT PRINTING OFFICE: 1985-544-130/10014 Region 4.