

Prepared in cooperation with the U.S. Environmental Protection Agency, Office of Ground Water and Drinking Water

# Occurrence of Selected Radionuclides in Ground Water Used for Drinking Water in the United States: A Reconnaissance Survey, 1998

Water-Resources Investigations Report 00-4273



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By Michael J. Focazio, Zoltan Szabo, Thomas F. Kraemer, Ann H. Mullin, Thomas H. Barringer, and Vincent T. dePaul

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### CONVERSION FACTORS, WATER-QUALITY UNITS, AND ABBREVIATIONS

Multiply	By	To Obtain
	Volume	
gallon (gal)	3.785	liter (L)
	3785	milliliter (mL)

#### Physical and Chemical Water-Quality Units and Common Abbreviations

**Temperature:** Water and air temperature are given in degrees Celsius (°C), which can be converted to degrees Fahrenheit (°F) by use of the following equation:

 $^{\circ}F = 1.8(^{\circ}C) + 32$ 

**Milligrams per liter (mg/L) or micrograms per liter (\mug/L):** Milligrams per liter is a unit expressing the concentration of chemical constituents in solution as weight (milligrams of solute per unit volume (liter)) of water. One thousand micrograms per liter is equivalent to one milligram per liter. For concentrations less than 7,000 mg/ L, the numerical value is the same as for concentrations in parts per million.

**Picocuries per liter (pCi/L):** Picocuries per liter is a unit expressing the concentration (or activity) of radionuclides in solution as particles emitted per unit volume (liter) of water. By definition, one gram of radium has one curie of activity. A picocurie is a millionth of a millionth of a curie. In practical terms, one picocurie per liter equals 2.2 radioactive disintegrations per minute per unit volume (liter) of water.

CSU, Combined Standard Uncertainty

MCL, Maximum Contaminant Level

MDC, Minimum Detectable Concentration

MRL, Minimum Reporting Level

NIRS, National Inorganics and Radionuclide Survey of the USEPA

USEPA, United States Environmental Protection Agency

## Occurrence of Selected Radionuclides in Ground Water Used for Drinking Water in the United States: A Targeted Reconnaissance Survey, 1998

By Michael J. Focazio, Zoltan Szabo, Thomas F. Kraemer, Ann H. Mullin, Thomas H. Barringer, and Vincent T. dePaul

#### ABSTRACT

The U.S. Geological Survey, in collaboration with the U.S. Environmental Protection Agency, the American Water Works Association, and the American Water Works Service Company, completed a targeted national reconnaissance survey of selected radionuclides in public ground-water supplies. Radionuclides analyzed included radium-224 (Ra-224), radium-226 (Ra-226), radium-228 (Ra-228), polonium-210 (Po-210) and lead-210 (Pb-210).

This U.S. Geological Survey reconnaissance survey focused intentionally on areas with known or suspected elevated concentrations of radium in ground water to determine if Ra-224 was also present in the areas where other isotopes of radium had previously been detected and to determine the co-occurrence characteristics of the three radium isotopes (Ra-224, Ra-226, and Ra-228) in those areas. Ninety-nine raw-water samples (before water treatment) were collected once over a 6-month period in 1998 and 1999 from wells (94 of which are used for public drinking water) in 27 States and 8 physiographic provinces. Twenty-one of the 99 samples exceeded the current U.S. Environmental Protection Agency drinking-water maximum contaminant level of 5 picocuries per liter (pCi/L) for combined radium (Ra-226 + Ra-228). Concentrations of Ra-224 were reported to exceed 1 pCi/L in 30 percent of the samples collected, with a maximum concentration of 73.6 pCi/L measured in water from a nontransient, noncommunity, public-supply well in Maryland. Radium-224 concentrations generally were higher than those of the other isotopes of radium. About 5 percent of the samples contained concentrations of Ra-224 greater than 10 pCi/L, whereas only 2 percent exceeded 10 pCi/L for either Ra-226 or Ra-228. Concentrations of Ra-226 greater than 1 pCi/L were reported in 33 percent of the samples, with a maximum concentration of 16.9 pCi/L measured in water from a public-supply well in Iowa. Concentrations of Ra-228 greater than 1 pCi/L were reported in 22 samples, with a maximum concentration of 72.3 pCi/L measured in water from a non-transient, noncommunity, public-supply well in Maryland.

Radium-224, which is a decay product of Ra-228 in the Th-232 decay series, was significantly correlated with Ra-228 (Spearman's rank correlation coefficient "r" equals 0.82) and to a lesser degree with Ra-226 (r equals 0.69), which is an isotope in the U-238 decay series. The rank correlation coefficient between Ra-226 and Ra-228 was 0.63. The high correlation between Ra-224 and Ra-228 concentrations and the corresponding isotopic ratios of the two (about 1:1 in 90 per-

cent of the samples) indicates that the two radionuclides occur in approximately equal concentrations in most ground water sampled. Thus, Ra-228 can be considered as a reasonable proxy indicator for the occurrence of Ra-224 in ground water.

The maximum concentration of Po-210 was 4.85 pCi/L and exceeded 1 pCi/L in only two samples. The maximum concentration of Pb-210 was 4.14 pCi/L, and about 10 percent of the samples exceeded 1 pCi/L. Areas with known, or suspected, elevated concentrations of polonium and lead were not targeted in this survey.

Three major implications are drawn for future radionuclide monitoring on the basis of this information: (1) grossalpha particle analyses of ground water should be done within about 48-72 hours after collection to determine the presence of the short-lived, alpha-particle emitting isotopes, such as Ra-224, which was detected in elevated concentrations in many of the samples collected for this survey; (2) the isotope ratios of Ra-224 to Ra-228 in ground water are variable on a national scale, but the two radioisotopes generally occur in ratios near 1:1, therefore, the more commonly measured Ra-228 can be used as an indicator of Ra-224 occurrence for some general purposes other than compliance; and (3) the isotopic ratios of Ra-226 to Ra-228 were less than 3:2 in many samples. These ratios corroborate results of previous studies that have shown the presence of Ra-228 (a beta-particle emitter) can cause a flaw in conventional radium compliance monitoring when the gross-alpha particle screen indicates no need for further analysis (the gross alpha particle activity is less than 5 pCi/L) even though the ratio of Ra-226 to Ra-228 is less than 3:2.

#### INTRODUCTION

The Drinking Water Initiative of the U.S. Geological Survey (USGS) provides scientific information for regulatory agencies and other water-resource planners and managers who must balance water-supply protection with the wise use of public funds (Patterson, 1997). In fulfilling the goals of the USGS Drinking Water Initiative, and to provide information on the occurrence of selected radionuclides in ground water, the USGS in collaboration with the U.S. Environmental Protection Agency (USEPA) performed a reconnaissance survey of selected radionuclides in ground water that is used as a source of drinking water in the United States.

Drinking-water standards for radionuclides have been in place since 1976 (U.S. Environmental Protection Agency, 1976). Revisions to the drinking water standards for radionuclides were proposed by the USEPA in 1991 but never enacted into law. The USEPA is currently (2000) taking action under a court agreement (Order on Stipulated Motion for Relief from Judgment or Order, and Motion to Dismiss All Consolidated Cases, Joseph L. Miller v. Carol M. Browner, Civ. Nos 89-6328 et al (D. Ore, 1996)) on the 1991 proposed revisions. This action includes establishment of a Maximum Contaminant Level (MCL) for uranium and either promulgating or stating the reasons for not promulgating any other elements of the proposal. As part of these actions, the USEPA published a Notice of Data Availability (NODA) in the Federal Register on April 21, 2000 (U.S. Environmental Protection Agency, 2000) to update the public and the regulated community on issues regarding radionuclides since the publication of the proposed radionuclides rule in 1991. The NODA summarizes the available information on the occurrence of radionuclides, among other things, and includes data from the 1998-99 reconnaissance survey reported in this report. Much of the data on occurrence of radionuclides in ground water summarized in the NODA were from previous occurrence surveys (Longtin, 1988); however, insufficient information on the occurrence of Ra-224 in water resources was available (Hess and others, 1985) until recently (Szabo and others, 2000).

The reconnaissance survey was designed to provide data and interpretative information on the potential national scope of the occurrence of the short-lived Ra-224 and its associated contribution to gross alpha-particle radiation in drinking-water supplies where it was presumed to exist in elevated concentration. In addition, the survey was designed to provide data and information on the co-occurrence characteristics of the three radium isotopes; Ra-224, Ra-226, and Ra-228. The survey focused on areas that have ground water with known, or suspected elevated concentrations of radium-224 (Ra-224; half life, 3.64 days), as inferred by previously published data and information on the occurrence of radium, geochemical models, geologic maps, and other geochemical or water-quality considerations.

Raw-water samples were collected once over a 6-month period in 1998 from 99 wells in 27 States and 8 physiographic provinces across the United States. Of these wells, 94 were used for public drinking-water supply, one was used for domestic supply; four research wells are not used for drinking-water supply. The samples, which were collected as close to the wellhead as possible, represent the raw (untreated) water that is used for drinking-water purposes. The public water-supply systems surveyed serve a broad range of population sizes, including several serving less than 500 people and one serving more than 1,000,000 people. The number of people that consume water from each well sampled is unknown.

#### **Purpose and Scope**

The purpose of this report is to provide information and data that describe concentration distributions of Ra-224 in drinking water from selected ground-water sources nationwide. Radiochemical analyses of raw, untreated ground water from public water-supply systems provide the source of the data. The USGS is providing these data to USEPA to augment existing sources of information to assess the potential national impacts of revisions to drinking-water standards (MCLs) and compliance monitoring for the targeted radionuclides. Results of this survey can also provide information needed to design a more complete survey of radium occurrence (particularly Ra-224) and co-occurrence (particularly of Ra-224, Ra-226, and Ra-228) in the Nation's ground-water resources.

#### Acknowledgments

The authors thank the public water-supply system personnel who voluntarily collected and shipped water samples to the respective laboratories. Daniel Pedersen, formerly of the American Water Works Association, and Daniel Schecter of the American Water Works Service Company provided crucial liaison support with the volunteers and helped select the sampling sites. David McCurdy, Ashok Banavli, and Jeffrey Raimondi of Duke Engineering and Services, and John Griggs and David Kappelman of USEPA, are among those who performed and helped coordinate the laboratory analyses. Lowell Ralston and Neal Nelson of USEPA and Patty Toccalino of the Oregon Graduate Institute provided guidance and review for the presentation of the human health risks of radium in drinking water. David Huber, William Labiosa, James Taft, and other members of the USEPA radionuclide rule assessment team, were instrumental in providing support, discussion, and suggestions for many aspects of the project from conception to completion. Alison Gordon, USGS, assisted in contacting volunteers and coordinating the sample collection. Glenn Patterson, USGS, helped in many aspects of the project from proposal development to reviews of parts of the manuscript. Robert Hirsch, USGS, provided useful comments on an early version of the manuscript. Colleague reviews were provided by Robert Kay, Lisa Senior, and David McCurdy.

#### General Background on Radionuclide Chemistry

There are about 2,000 known radionuclides, which are species of atoms that emit radiation as they undergo radioactive decay through emission of alpha particles, beta particles, or gamma rays. Naturally occuring radionuclides are ubiquitous trace elements found in rocks and soils and, in general, radionuclides can be categorized in three ways:

- by type of radioactive decay (alpha, beta, or gamma emission),
- by radioactive decay series,
- as naturally occurring or manmade,

Some of the naturally occurring and manmade radionuclides are directly regulated by the current drinking-water standards. The natural radionuclides include the primordial elements that were incorporated into the earth's crust during its formation, the radioactive decay products (or progeny) of these primordial elements, and radionuclides that are formed in the atmosphere by cosmic ray interactions. Manmade radionuclides are produced through the use of nuclear fuels, radiopharmaceuticals, and other activities of the nuclear industry and have been released into the atmosphere as the result of atmospheric testing of nuclear weapons and, in rare cases, accidents at nuclear-fuel stations, and discharge of radiopharmaceuticals. Further discussion on manmade radio-nuclides is beyond the scope of this report.

The two types of isotopes with radioactive decay that carry the most health risk due to ingestion of water are alpha-particle emitters and beta/photon-particle emitters (Lappenbusch and Cothern, 1985). Many radionuclides are mixed emitters with either an alpha or beta emission coupled with gamma (photon) emission, or in some cases, all three. Each radionuclide has a primary mode of disintegration. The most common, heavy, naturally occurring radionuclides are largely alpha-particle emitters, although their progeny often emit beta particles. The alpha-particle emitting radionuclides discussed in this report include two isotopes of radium (Ra-224 and Ra-226) and polonium-210. Alpha radiation is composed of a particle, consisting of two protons and two neutrons, spontaneously emitted from the nucleus of a subset of radioactive elements (mostly the heaviest elements) during radioactive decay. Alpha radiation is ionizing radiation, meaning that it strips electrons from adjacent atoms as it passes. Alpha radiation cannot penetrate skin; thus, an alpha-particle emitting radionuclide must be ingested in order to come into contact with internal tissue. Because of the large size, alpha particles are likely to collide with cell tissue, causing tissue damage. An accumulation of tissue damage in the cell nucleus may lead to cell mutation and potential cancer.

The naturally occurring radionuclides derived from uranium-238, thorium-232, and uranium-235 are products of the radioactive decay series known as the uranium, thorium and actinium series, respectively. Each decay series follows a known sequence of radioactive decay, producing various isotopes that also emit either an alpha or a beta particle as they decay (fig. 1). Each series terminates with a stable isotope of lead. The crustal abundance of U-235 is very low in comparison with the other decay series (U-238 to U-235 mass ratio is on the order of 140 to 1). Thus, the actinium series is not considered an important source of radionuclides in drinking water derived from ground water and is, therefore, not included in figure 1.

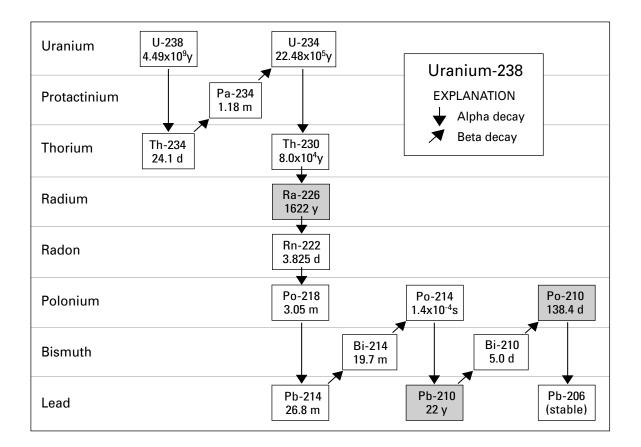
Radium in nature is composed of four isotopes: Ra-223, Ra-224, Ra-226, and Ra-228. Radium-223 is a member of the U-235 decay series and, therefore, rarely occurs in the environment in high concentrations. Radium-224 is the fifth member of the Th-232 decay series, has a half-life of 3.64 days, and decays by alpha-particle emission. Radium-226 is the fifth member of the U-238 decay series, has a half-life of about 1,602 years, and decays by alpha-particle emission. Radium-226 is the most abundant radium isotope in the environment in terms of actual mass because of its long halflife. Radium-228 is the second member of the Th-232 decay series, has a half life of 5.75 years, and decays by beta-particle emission.

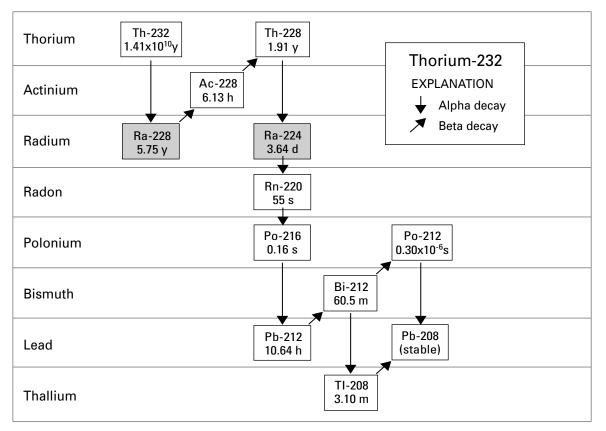
The occurrence of radionuclides in ground water depends first on the presence and solubility of the parent element. Each radioactive decay product has unique chemical characteristics common to the element but differing slightly by isotope. For the general purposes of most occurrence analyses, the different isotopes of radium are chemically indistinguishable. The occurrence of a parent radionuclide in solution does not necessarily indicate the presence of its decay products. For example, U-238 (parent product of Ra-226) tends to be the least mobile in oxygen-poor ground water, and tends to be strongly adsorbed onto humic substances. Conversely, radium tends to be most mobile in oxygen-poor ground water that is chloride-rich with high concentrations of total dissolved solids (Kraemer and Reid, 1984; Zapecza and Szabo, 1987). Radium chemically reacts similarly to other divalent alkaline-earth cations such as calcium, strontium, and barium. Therefore, in aquifers with limited sorption or ion-exchange sites, radium solubility can be enhanced by the common-ion effect where competing cations are present in abundance. Recently, high concentrations of radium were found to be associated with ground water that was geochemically affected by land use (agricultural) practices in recharge areas, presumably due to waters enriched with competing ions such as hydrogen, calcium, and magnesium (Szabo and dePaul, 1998). Ra-228 was detected in about equivalent concentrations as Ra-226 in an acidic aquifer in New Jersey, where agricultural fertilizers were ruled out as a source of radium (Szabo and dePaul, 1998).

When an alpha particle is ejected from the nucleus of a radionuclide during decay, the newly created progeny radionuclide recoils in the opposite direction. This process is known as the alpha-recoil process, and it enhances the solubility of alpha-particle emitting radionuclides. The energy associated with this recoil is  $10^4$ – $10^6$  times larger than typical chemical-bond energies (Cothern and Rebers, 1990) and can cause atoms on the surface of a grain to be recoiled directly into the water in pore spaces.

#### **Existing Sources of Information for Radionuclide Data**

Results of the National Inorganics and Radionuclides Survey (NIRS) (Longtin, 1988) were used to develop the USEPA proposed radionuclide rule in 1991 (Federal Register Vol. 56, No. 138). The objective of the NIRS was to characterize the occurrence of a variety of constituents present in community ground-water supplies (finished water) in the United States, its territories, and its possessions. The survey included a random sample from 990 collection sites at wellheads and entry points to water-distribution systems that were stratified into four population categories that represented about 2 percent of drinking-water supplies in each category. Longtin (1988) summarized the NIRS data and compared it with data from other studies completed at that time, especially that of Hess and others (1985), who summarized existing data from national and State surveys to describe the occurrence of radioactivity in public water supplies in the United States. Zapecza and Szabo (1987) summarized the occurrence data for uranium, radium, and radon in ground water in the United States on the basis of USGS, State regulator, and university research documents and databases. A comparable national survey for radionu-





**Figure 1**. Diagram showing uranium-238 and thorium-232 radioactive decay series. (Radionuclides of interset in this study are shaded). [Times shown are half-lives: y, years; d, days; h, hours; m, minutes; s,seconds]. (From Hall and others, 1985.)

clides has not been done since the cited examples were completed in the late 1980's.

#### **Radium in Water Resources of the United States**

As previously noted, natural radium is composed of four isotopes, with three of the four (Ra-224, Ra-226, and Ra-228) to be discussed in this report. To date, measured radium in surface waters has been traced to discharge from ground water (Elsinger and Moore, 1983) or industrial waste (Kraemer and Curwick, 1991); discussion in this report is thus limited to ground water.

#### **Occurrence of Radium-224**

The NIRS (Longtin, 1988) did not include Ra-224. Based on a limited number of analyses, however, Hess and others (1985) report that the activity of Ra-224 can be equal to or greater than the activity of Ra-228 in ground water; they assume concentrations could reach as high as 30 to 40 pCi/L. Hess and others (1985) conclude that Ra-224 activity is likely unsupported because activities of its parent, Th-228, are usually less than 0.01 pCi/L. Ra-224 can enter ground water by alpha recoil during the decay of Th-228 adsorbed on the surface of aquifer solids. Elsinger and Moore (1983) used radium to trace ground-water discharge in the South Carolina Coastal Plain and documented the occurrence of Ra-224 in the local ground-water resource.

#### **Occurrence of Radium-226**

Ra-226 is a progeny of U-238, and U-238 is commonly found in ground water. The relatively high solubility of U-238 as compared to thorium is a cause of its widespread distribution, and, thus, the widespread distribution of its daughter products such as Ra-226. The geochemical properties of Ra-226 differ from those of U-238, however, and co-occurrence is not common (Szabo and Zapecza, 1991) because the degree and chemical conditions of mobilization of the parent U-238 and product Ra-226 are different. Uranium forms soluble complexes particularly with carbonates under oxygen-rich conditions, and precipitates from ground water under oxygen-poor conditions (Zapecza and Szabo, 1987; Cothern and Rebers, 1990). The concentration of uranium in water and rock can thus be expected to range widely with different geochemical environments. Szabo and Zapecza (1991) detail the differences in the occurrence of uranium and Ra-226 in oxygen-rich and oxygen-poor waters of hard rock aquifers.

The most extensive region in the Nation where Ra-226 occurs in elevated concentrations in ground water is in the north-central states, including southern Minnesota, Wisconsin, northern Illinois, Iowa, and Missouri (Zapecza and Szabo, 1987; Gilkeson and others, 1983; Kay, 1999). In these States, the drinking-water wells tap deep aquifers of Cambrian and Ordovician sandstones and dolomites and Cretaceous sandstones. These aquifers tend to have limited sorption sites, and radium solubility is enhanced by the common-ion effect wherever the concentration of total dissolved solids are high (Gilkeson and others, 1983). In some areas, such as northern Illinois, reduction of sulfate decreases

coprecipitation of Ra-226 with barium sulfate, another mechanism which, if present, tends to limit dissolved Ra-226 (Gilkeson, and others, 1983).

Ra-226 is also found in high concentrations in water derived from aquifers that straddle the Fall Line of the southeastern States from Georgia to New Jersey (King and others, 1982; Zapecza and Szabo, 1987; Szabo and dePaul, 1988). These aquifers are composed of unconsolidated sands that contain fragments of uranium-bearing minerals derived from the crystalline rocks of the Blue Ridge and Piedmont provinces. Sands of the Coastal Plain where uranium and radium concentrations tend to be the highest (Zapecza and Szabo, 1986) were directly derived from these crystalline rocks as fluvial deposits. Furthermore, in areas with saltwater intrusion or brackish water, the common-ion effect again is likely the cause for high concentrations of dissolved Ra-226 (Kraemer and Reid, 1984; Miller and Sutcliffe, 1985).

Of the sites surveyed for NIRS, activities of Ra-226 equaled or exceeded the MRL of 0.18 pCi/L in 40.2 percent of the samples (Longtin, 1988). The national median activity (of the samples with concentrations greater than 0.18 pCi/L) was 0.39 pCi/L and the maximum was 15.1 pCi/L. About 1 percent of the samples had concentrations greater than 5 pCi/ L, the value that would place the drinking-water supply system out of compliance with the MCL for radium, even in the absence of Ra-228.

#### **Occurrence of Radium-228**

Although Ra-228 is chemically similar to Ra-226, its distribution in ground water can be different for several reasons. The relatively short half-life of Ra-228 limits the potential for unsupported transport (transport without the presence of the parent product in solution) relative to that of the longer lived Ra-226 isotope. Consequently, because of the relatively slow movement of ground water in most aquifers, Ra-228 cannot migrate far from its source before it decays into its progeny. Thorium-232, the parent of Ra-228, is extremely insoluble (Cothern and Rebers, 1990) and is not subject to mobilization in most ground-water environments. The insolubility of thorium (unlike the moderate solubility of uranium) thus limits the distribution of Ra-228 in ground water to some degree.

Michel and Cothern (1986) developed a national model to depict the occurrence of Ra-228, based primarily on the thorium content of the rock or sediment material that composed the aquifer. Data from NIRS showed that Ra-228 activities tend to be the highest in arkosic sand and sandstone aquifers, indicating the model is representative of the distribution of Ra-228. Generally, the areas that are associated with the highest potential for Ra-228 include the Coastal Plain aquifers that straddle the Fall Line from Georgia to New Jersey and Pennsylvania. Ground-water data from this region confirms the presence of dissolved Ra-228 in Coastal Plain and Paleozoic quartzite (metamorphosed arkosic sandstone) aquifers (Szabo and dePaul, 1998; King and others, 1982; Senior and Vogel, 1995). Large areas of northern Illinois, Iowa, Minnesota, and Wisconsin were also ranked as having a high potential for the occurrence of Ra-228 as a result of the presence of sandstone aquifers with high total dissolved solids; actual occurrence data verifies this distribution (Gilkenson and others, 1983). In contrast, aquifers that were mostly composed of alluvial or glacial sand and gravel deposits were rated as having low potential for Ra-228 occurrence in those same states.

The High Plains aquifers and feldspathic sandstone aquifers were also considered to have some potential for Ra-228 occurrence. Granitic rocks that have the prerequisite geochemical characteristics for Ra-228 occurrence underlie other parts of the country such as local areas in Colorado, Montana, and California. Aquifers in these rock types do not yield much water and are not used extensively for large public water supplies. Localized alluvial valley sandstone aquifers in these same areas may also contain dissolved Ra-228. These are areas that require additional testing in the future.

The concentration of Ra-228 was greater than 1 pCi/L at about 12 percent of the sites sampled for NIRS (Longtin, 1988). The median activity of Ra-228 for samples with concentrations greater than 1 pCi/L (the MRL) was 1.47 pCi/L, and the maximum was 12.1 pCi/L. About 18 percent of the samples had concentrations that exceeded 2 pCi/L, the concentration that would put a system out of compliance if it had a Ra-226 concentration of 3 pCi/L (the concentration of Ra-226 that triggers analysis for Ra-228). Four percent of the samples had concentrations that exceeded 5 pCi/L, the value that would place the drinking-water-supply system out of compliance with the MCL for radium even in the absence of Ra-228.

#### Other Naturally Occurring Alpha-Particle Emitting Radionuclides

Generally, the alpha-particle emitting radionuclides of concern for drinking-water standards include those with halflives that are considered long enough to travel through drinking-water distributions systems and be ingested. Radionuclides having a half-life of 1 hour or less were not considered in the group proposed for regulation in the 1991 proposed rule (U.S. Environmental Protection Agency, 1991) because most are likely to decay before the drinking water is consumed. The widespread occurrence of Ra-224 in New Jersey (Szabo and others, 1998a) increased concerns regarding the occurrence of radionuclides with half-lives on the order of 1 day or a few days in drinking-water supplies. Alpha-particle emitting radionuclides with a half-life longer than 1 hour include:

Decay series	Alpha-particle emitting radionuclides with half-life longer than 1 hour
Uranium	U-238, U-234, Th-230, Ra-226, Rn-222, Po-210
Actimium	U-235, Po-231, Th-227, Ra-223
Thorium	Th-232, Th-228, Ra-224, Pb-212

Long-lived (half-life greater than 1 day) beta-particle emitting radionuclides, other than Ra-228 and Pb-210 are, in large part, found in water resources affected by manmade sources of radiation. The naturally occurring Ra-228 and Pb-210 are the only beta-particle emitting radionuclides analyzed in this study. Other radionuclides, such as the thorium isotopes, were analyzed in this study for quality-control purposes only.

Hess and others (1985) summarized the limited data on Po-210 concentrations in ground water. Po-210 is particle reactive and is not expected to be found in high concentrations in most natural waters. Po-210 was found at an unusually high concentration (290 to 607 pCi/L) in a well used for drinking water in Louisiana (Mullin, 1982) and was above 2 pCi/L in some wells in the Grants Mineral Belt of New Mexico (Kaufmann and others, 1976). The source of Po-210 in the well in Louisiana is unknown. Uranium ore of mineable grade was presumed to be the source of Po-210 in the wells in New Mexico. It is not known if changes in water quality as a result of mining increased the mobility of the Po-210. Po-210 has also been found in a shallow aquifer in west-central Florida (Harada and others, 1989). Uranium-enriched phosphatic strata that contains sulfidic acidic water high in Rn-222 characterize the aquifer. Water of this geochemical type is rare in the environment and not typical of water used for drinking-water supplies.

The geochemistry and occurrence of Rn-222 and all other radionuclides not specifically mentioned above are not within the scope of this study. A separate drinking-water standard for Rn-222 has been proposed by USEPA (1999b).

#### **Limitations of Data Sources**

The analytes quantified by NIRS included Ra-226, Ra-228, Rn-222, uranium (mass analysis), and gross alphaparticle and gross beta-particle activities. The gross alphaand gross beta-particle activities were surveyed, in part, due to recognition of the potential combined contribution to radioactivity in drinking water from the many other naturally occurring and manmade radioisotopes. The gross alpha- and beta-particle activities were determined after variable holding times and, therefore, do not give complete information about the contribution of short-lived radionuclides. Radium-224 was not analyzed in NIRS or any other national survey, and since the completion of NIRS, no national-scale survey to characterize radium in drinking water has been done.

Recent investigations to determine more accurately the distribution of radioactivity in selected aquifers in New Jersey led to the finding that gross alpha-particle activity varies considerably based on the amount of time that passed between sample collection and sample analysis (Szabo and others, 1998a; Parsa, 1998). Ongoing scientific study has shown that this phenomenon is the result of the presence of short-lived radionuclides that previously were not believed to be present in large concentrations in ground water. Modifications and improvements to analytical techniques led to recognition that large amounts of short-lived Ra-224 were widespread in the water of the targeted aquifer (Szabo and others, 1998b).

The New Jersey study illustrates that targeted, national-scale radionuclide data from drinking-water sources is incomplete and insufficient for assessing the occurrence of short-lived radionuclides, such as Ra-224, and their contribution to gross-alpha particle activity.

# DATA COLLECTION AND LABORATORY ANALYTICAL METHODS

Samples were collected from wells selected for the purposes of assessing the occurrence of radium-224 in areas known, or inferred to be associated with, high concentrations of radium in drinking water supplies. Targeted sampling makes use of knowledge about the likely controlling factors on water quality in particular areas and can be useful in identifying water-quality problems that would be missed or underrated by typical random survey designs (Alley, 1993). In addition, targeted sampling designs provides data to test specific hypotheses. For example, hypotheses regarding the relation of the occurrence of radium with geologic and other factors, the ratios of isotopes of radium in water, and factors controlling the presence of the short-lived Ra-224 can be tested with these data. Finally, this approach provided a way to maximize limited resources in order to quickly assess the potential scope of the occurrence of elevated concentrations of these radionuclides in drinking-water sources on a national scale and determine whether further study is warranted.

One well was sampled once from each public watersupply system (except those used for quality control where sequential split samples were taken). Consequently, not accounted for are variability in concentrations of radionuclides as a result of long-term and seasonal changes or other factors associated with well construction and the operations of the public water-supply systems (Alley, 1993; Alley and Cohen, 1991; Reilly and others, 1989). A single sample thus may not necessarily represent the water quality of the groundwater resource used by the public water-supply system over a long period of time. An individual sample is representative of the quality of the local ground-water flow that contributes water to the targeted well for the time at which it was sampled. This sample may not represent the overall water quality of the associated ground-water resource used by the public water-supply system or ultimately delivered to their customers, particularly where waters from various wells or sources are mixed. Individual samples, which provide a snapshot of water quality at the time of sampling, can provide reasonable depictions of the general water quality of an aquifer, however, particularly where slow movement of ground water limits rapid fluctuations in water chemistry.

#### Well Selection

Areas in 27 States were targeted for sampling. These selections were based on availability of previous occurrence data and models, geologic maps, and known or inferred geochemical properties of aquifers (Hess and others, 1985; Michel and Cothern, 1986; Zapecza and Szabo, 1987; Longtin, 1988). The well selections were narrowed to targeted areas with known, or suspected, high concentrations of radium in the ground water within the same physiographic province. Wells from public water-supply systems in these areas were randomly selected for sampling by personnel from the American Water Works Association (AWWA) and American Water Works Service Company (AWWSC) (fig. 2). At least one sample was collected from eight of the major physiographic provinces in the United States.

The water purveyors selected to participate were each asked to complete a questionnaire to provide information on well age and construction, local geology, and the population served by their system (Appendix 1). Data on the population served by the surveyed public water-supply systems represent both quantified and estimated numbers. Estimates made by the water purveyors typically represent the maximum population that their system serves.

#### Sample-Collection Methodology

Unfiltered, raw-water samples were collected as close to the wellhead as possible after the pump had been running a minimum of one-half hour. Latex gloves were worn during sample collection to avoid sample-handling contamination. Samples were shipped overnight to the laboratory. Chemical separation and analysis was begun within 48 hours for Ra-224, Ra-226, and Po-210, and as soon as possible after that for Ra-228 and Pb-210. The samples were acidified to pH 2 with reagent-grade nitric acid (HNO<sub>2</sub>) and filtered with a 0.45-micrometer plate filter. Analysis for concentrations of Th-232, Th-230, and Th-228 were conducted if the Ra-224 concentration was 4 pCi/L or greater. Concentrations of alpha-particle emitting Ra-223 were determined for a randomly chosen subset of samples. Results from various analytical methods were compared and extensive evaluation of quality-assurance samples was completed .

#### Analytical Methodology

Measurement of radioactivity determines the amount of radioactivity in a given volume of water. For water, the amount of radioactivity generally is expressed in units of picocuries per liter. A picocurie per liter equals 2.22 radioactive disintegrations per minute per liter of water. Many techniques are available for measuring radioactivity; the specific technique chosen depends on the radionuclide whose concentration is being determined. Alpha spectrometry, gamma spectrometry, and low-background gas-proportional beta counting were the general techniques used in this study (table 1); the specific details of each analysis are detailed.

Alpha spectrometry specifies the amount of alpha radiation emitted at specified energy levels, thus allowing quantitation of individual radionuclide concentrations. Solidstate detectors, such as germanium or passivated ion-implanted planar silicon (pips) detectors, are very sensitive and the most commonly used sensors. Gamma spectrometry specifies the amount of gamma radiation emitted at specified energy levels, thus allowing quantitation of individual radionuclide concentrations. Beta-particle activity is counted in a low-background, gas proportional counter. Detection efficiency is highest for beta particles emitted with the highest energy.



Figure 2. Major physiographic provinces of the United States and locations of wells with completed radium-224 analysis, 1998.

Alpha spectrometry was the primary method used to determine the concentration of alpha-particle emitting radionuclides of radium (Ra-224 and Ra-226). Improvements in separation and purification chemistry (Sill and Williams, 1981) have allowed the alpha spectrometry measurement technique to become available as a rapid, accurate, low-cost analytical method for the alpha-particle emitting radionuclides of radium. The counting time required for alpha spectrometric determination of the radium concentrations is shorter, and the instrumentation is not as bulky as that required for gamma-spectrometric determination; thus, many analyses could be performed quickly to obtain timely results for this large-scale survey.

The alpha-spectrometry method used and briefly described here is not yet approved by either USEPA or the American Society for Testing Materials (ASTM) but is only slightly modified from the widely used method of Sill and others (1979). After acidification and filtration, a radioactive barium (Ba) radionuclide (Ba-133) was added as a yield tracer. Cation exchange chromatography (Bio-Rad AG 50W-X8 cation exchange resin) was used to separate and purify the Ra and Ba. The eluent from the exchange resin was removed with 8 molar HNO<sub>3</sub>, and the Ra and Ba were co-precipitated with barite (BaSO<sub>4</sub>) in a cold-water bath using a seeding suspension (Sill and Williams, 1981). The seeding suspension is needed to ensure the formation of uniform fine-grained crystals of the BaSO<sub>4</sub> that were then mounted directly on filter paper, which was counted for 100 minutes using alpha spectrometry with adjusted instrument operating conditions that minimize background contamination (Sill and Olsen, 1970). The count time was chosen to ensure a contractual minimum detectable concentration (defined in following section) of 1 pCi/L or less for most samples. Yield was determined by gamma-spectrometric analysis of the concentration of the radioactive Ba-133 yield tracer.

The concentration of the beta-particle-emitting Ra-228 radionuclide was determined on a separate aliquot using the USEPA-approved low-background beta count of the actinium-228 progeny (Krieger and Whittaker, 1980) because the concentration of this radionuclide could not be determined using alpha spectrometry. The beta-counting technique provided a contractual minimum detectable concentration (defined in following section) of about 1 pCi/L for Ra-228.

Radium concentrations in about one-third of the

samples were also determined using gamma spectrometry as part of the quality-assurance program. Analysis of the concentration of the beta-particle emitting Ra-228 isotope can be performed on the same sub-sample aliquot as the alpha-particle-emitting Ra-226 and Ra-224 isotopes.

Several approaches to gamma-spectrometric measurement of Ra-224 concentrations have been compared and found to give consistent results (Szabo and others, 1998b); the gamma-spectrometric analytical technique used and briefly described here is not yet approved by either USEPA or the ASTM and is only slightly modified from the original method of Elsinger and others (1982). After acidification and filtration, Ra was coprecipitated with BaSO, and mounted directly on filter paper. The Ra-224 activity was counted using the 238.6 KeV energy emission of the Pb-212 progeny for 1,000 minutes 52 hours after coprecipitation using a lead-shielded high-efficiency (about 40 percent) gamma spectrometer with a well-type germanium-lithium detector. The radium-bearing BaSO<sub>4</sub> was recounted for 1,000 minutes about 28 days later to determine the concentration of Ra-226 and Ra-228. The count time was chosen to ensure a contractual minimum detectable concentration of 1.0 pCi/L for each radionuclide. The barium yield was determined gravimetrically.

The concentration of the alpha-particle-emitting Po-210 radionuclide was determined using alpha-spectrometry. The sample bottle was soaked for 1 hour in concentrated  $HNO_3$  to remove adsorbed Po-210 from the container walls. The Po-210 was deposited from a concentrated hydrochloric acid (HCl) solution onto a polished copper disk. Sample counting time by alpha spectrometry was 100 minutes to ensure a contractual minimum detectable concentration of about 1 pCi/L. Special care was taken to minimize counter contamination for the volatile polonium radionuclides using the techniques

outlined by Sill and Olsen (1970). The concentration of a Po-208 radioactive isotope tracer added to the sample was determined to compute yield.

The concentration of the beta-particle-emitting Pb-210 radionuclide was determined using the USEPA-approved low-background beta count of the bismuth-210 progeny after 10-day ingrowth. The Pb-210 was preconcentrated and purified using cation exchange chromatography (I-Chrome Pb-specific cation exchange resin). Sample counting time was 300 minutes to ensure a contractual minimum detectable concentration of about 1 pCi/L.

The concentration of the alpha-particle-emitting radionuclides of thorium (Th-232, Th-230, and Th-228) were determined using alpha-spectrometry on a selected number of samples. The concentration of the beta-particle-emitting Th-234 isotope, a member of the U-238 decay series, cannot be determined using this technique. Because Ra-224 is a direct progeny of Th-228 originating in a different decay series (the Th-232 decay series) than Th-234, the measurement of Th-234 concentrations was not necessary. The isotopes of thorium were preconcentrated and purified using cation-anion exchange chromatography (Bio-Rad AG 1-X8 anion exchange resin) from which the Th isotopes were eluted with dilute HCl, then electroplated at 1 ampere for 60 minutes onto a stainless-steel disk. Sample counting time by alpha spectrometry was 100 minutes to ensure a contractual minimum detectable concentration of about 1 pCi/L.

#### Reporting of Analytical Results and Statistical Calculations with Raw-Data Values

Raw, unrounded values for all radionuclide concentrations were reported directly by the laboratory, along with the minimum detectable concentration (MDC), and a combined standard uncertainty (CSU) or a counting error. An explanation

**Table 1.** Radionuclides analyzed in the United States Geological Survey reconnaissance survey, associated analytical method, and the number of samples (including environmental and quality-control samples) analyzed by each method [—, not applicable]

Methods										
Radionuclide	Alpha spectrometry	Number of samples	Gamma spectrometry	Number of samples	Beta counting	Number of samples	Alpha and gamma spectrometry	Number of samples Split with both methods Split with both methods in duplicate	Beta counting and gamma spectrometry	Number of samples Split with both methods Split with both methods in duplicate
Ra-224	Х	90	х	12	_	_	Х	15 36	_	_
Ra-226	x	90	Х	12	_	_	х	15 36	_	_
Ra-228	-	_	x	12	Х	90	—	—	х	15 36
Pb-210	_		_	_	Х	109	_	_	_	_
Po-210	Х	113	-	_	_	_	—	_	—	—
Thorium	Х	33			—		—			_

of these terms is provided in this section.

The decay of any radionuclide occurs at a given rate over a specific period of time. In any instant of time, however, the rate of radiation is random, resulting in a degree of uncertainty in the measurement known as the counting uncertainty term (also called the counting error). Thus, any measurement of radioactivity has a counting uncertainty independent of, and in addition to, laboratory sources of analytical uncertainty. Increasing the time of the count can decrease the counting uncertainty term. The greater the degree of uncertainty, the more difficult it is to quantify small amounts of radiation. Quantifying small amounts of radiation is further complicated because the analytical instruments contain background radiation that varies with every measurement. The radiation from the radionuclide being quantified must be clearly distinguishable above the ambient background radiation. A contractual minimum detectable concentration (American Society of Testing Materials, 1999) is contractually determined with the laboratory before a project begins and is generally based on the scope of the project and available resources. The contractual (also called the *a priori*) MDC was targeted for 1 pCi/L for all radionuclides analyzed in this study. The sample-specific MDC is computed individually for each radionuclide analysis on the basis of instrument operating conditions at the time of measurement and reported by the laboratory for each analysis. For purposes of this paper, the sample-specific (or *a posteriori*) MDC will be referred to simply as the MDC unless otherwise noted.

Uncertainty can be reported in a variety of ways; the most common include (1) the sum of the laboratory and counting uncertainty, known as the combined standard uncertainty (CSU, also called the total propagated uncertainty), and (2) a counting uncertainty only. There is a 67-percent or 95-percent probability (based on one or two standard deviations, respectively) that the true value of the radionuclide concentration is within the measured value plus or minus the uncertainty. The uncertainty terms are generally smaller than the measured value except when the measured values are low concentrations (near the MDC). The uncertainties associated with the various radionuclides in this report are provided directly by the individual laboratories as:

- One standard deviation of the CSU: Ra-224 and Ra-226 by alpha spectrometry.
- Two standard deviations of the CSU: Ra-228 and Pb-210 by beta counting, Po-210 by alpha spectrometry.
- One standard deviation of the CSU: Ra-224, Ra-226, and Ra-228 by gamma spectrometry.

The raw analytical result is interpreted in the context of the CSU and the MDC. The raw result specifies the instrumental reading after the count is completed, even if it is a negative value. A negative value is a result from a radiation measurement (count) of a sample over a fixed interval of time where the measured radioactivity from the sample is less than the long-term average background radiation determined routinely by the instrument. A negative result simply indicates that it is unlikely the radionuclide is present in a quantifiable amount.

Table 2. Number of public water-supply systems sampled,
by range of estimated number of people served
[ >, greater than]

Range of estimated number of people served	Number of public water-supply systems sampled
0–500	8
501-1,000	7
1,001–3,300	21
3,301–10,000	29
10,001–100,000	22
100,001-1,000,000	5
> 1,000,000	1
	$\Sigma$ 93 wells

Note: Two public water-supply systems did not provide information, and four wells surveyed were not affiliated with public water supplies.

Raw, unrounded values of concentrations reported directly by the laboratories were used to compute standard distributional statistics and correlation coefficients for all radionuclides. Correlations were calculated using the Spearman correlation coefficient. The Spearman correlation coefficient (correlation of the ranks of the data, not the actual values) is a measure of the degree to which the two data sets increase (or decrease) together. Negative values were set to zero for computational, but not reporting, purposes. No other manipulation or screening of the raw-data values was attempted unless otherwise stated in order to avoid biases due to omitting the low but detectable concentrations. Consequently, some calculations were performed with raw data that were at or below the MDC and (or) the CSU in a limited number of cases. This approach is a simple way to deal with the various possible combinations of raw values and associated CSU and MDC. This approach is not expected to affect the majority of data above 1 pCi/L, the contractual (a priori) MDC targeted by the laboratories. For example, ratios of radionuclide concentrations were determined using raw values; it is understood, however, that ratios may have bias when concentrations are below the MDC and (or) the CSU.

Statistical, or other calculations that omit data reported below the MDC and CSU are also biased, however, because some of the values omitted may actually be detectable radioactivity. Further, the omitted data still represent measurements of environmental radioactivity at levels that cannot be quantified; their omission biases the overall population distribution. Alternative approaches to calculating summary, and other statistics on water-quality data with concentrations near or below quantitation or reporting levels were discussed by Helsel (1990) and, although not used in this report, provide additional insights. These approaches are typically designed for concentrations that are reported as "less than" a minimum value, usually a minimum reporting level (MRL), whereas the raw unrounded data values were reported by the laboratories for the present study. For example, one such approach provides that a numeric value one half the MRL concentration be substituted for the sample result before summary statistics are calculated. The latter approach has been shown to be a more appropriate representation of the entire distribution of data below and above the MRL than simply ignoring the samples with concentrations below the MRL. The lack of a documented comparison of alternative approaches for radionuclide data preclude arbitrary use of such simple solutions that accurately portray the entire distribution of data for this study. Consequently, the correlations and other calculations that were performed with all data regardless of the associated MDC or CSU contain an unknown, but small, amount of uncertainty and potential bias, mostly limited to concentrations less than about 1 pCi/L (the contractual MDC). Attempts to quantify this uncertainty were beyond the scope of this report.

#### OCCURRENCE OF SELECTED RADIONUCLIDES IN GROUND WATER USED FOR DRINKING WATER

A total of 94 wells from public water supplies were sampled, as were 1 domestic well and 4 research wells (fig. 2). The populations served by the public water-supply systems that participated in the reconnaissance survey range from less than 500 to more than 1,000,000 (table 2). The public water-supply systems selected for the survey incorporate most areas depicted by Michel and Cothern (1986) as those where a medium to high relative probability of elevated concentrations of Ra-228 were expected to occur in ground water. Some of the areas in the western United States ranked with medium-to-high probability of containing elevated dissolved Ra-228 in ground water by Michel and Cothern (1986) were not included in this survey because large populations rarely rely on public ground-water sources in those areas. Because of the limited resources available to the study, small systems relying on ground water in the western United States were generally not sampled. We note this is an issue that needs to be resolved in future sampling work. The estimates of populations served are provided here for a general perspective on the distribution of public water-supply systems surveyed; the number of people that consume water from each well sampled is unknown. Accordingly, no attempt was made to analyze or associate the occurrence data on the basis of population served by public water-supply systems.

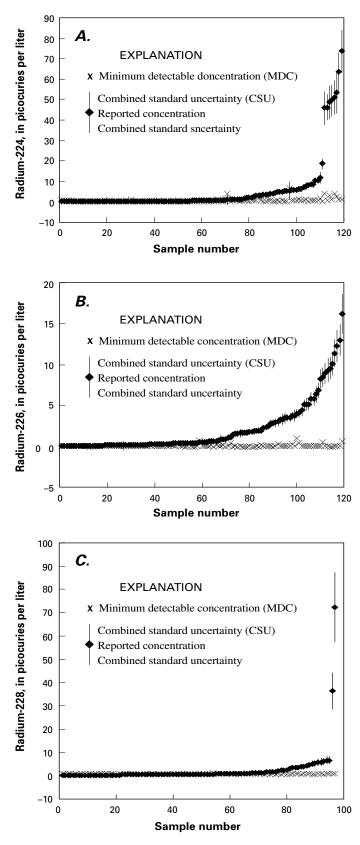
Twenty-one of the 99 samples exceeded the current combined radium standard. The reported concentrations of each radionuclide and the reported concentrations plus or minus the CSU were compared to the MDC (table 3). The lesser amount of uncertainty that is associated with the Ra-226 analyses than with the Ra-224 or Ra-228 analyses results in a higher percentage of samples with concentrations and CSUs that are greater than the MDC in the Ra-226 samples than in the other isotopes. Fifty-four percent of the reported Ra-224 concentrations were greater than the MDC; 88 percent of the Ra-226 concentrations were greater than the MDC, and 40 percent of the Ra-228 concentrations were greater than the MDC. This relative difference in analytical uncertainty is further illustrated by comparing the cumulative frequency of the reported concentrations, the concentrations plus and minus the CSU, and the MDC for each isotope (fig. 3). Table 3 and figure 3 provide insight and perspective on the reported concentrations and the associate uncertainties in the data base. Additional analyses of the relative uncertainties is beyond the scope of this report. Forty-five percent of the Po-210 concentrations were greater than the MDC and 36 percent of the Pb-210 concentrations were greater than the MDC. The distributions of the concentrations of all radionuclides analyzed are summarized in tables 4 and 5, and radium isotope co-occurrence is statistically summarized by analysis of correlation in table 6. Other radionuclides analyzed, such as thorium isotopes and Ra-223, are strictly part of the quality-assurance analysis and to test the experimental methods and are not within the scope of this discussion. The data for all radionuclides analyzed, however, are presented in Appendix 1.

#### Radium-224

Concentrations of Ra-224 in samples analyzed for the survey ranged from less than the MDC to 73.6 pCi/L (table 4). The Ra-224 concentration of 73.6 pCi/L is the

**Table 3.** Percentage of samples with reported concentrations that are greater or less than the minimum detectableconcentration (MDC) plus or minus the combined standard uncertainty (CSU) for all radium isotopes analyses(MDC< minimum detectable concentration; CSU, combined standard uncertainty)</td>

	Percentage of sample (including duplicate) results with						
Radionuclide	than the MDC and the than the MDC and the thar concentration minus the concent		The reported concentration less than the MDC and the concentration plus the CSU greater than the MDC	The reported concentration less than the MDC and the concentration plus the CSU less than the MDC			
Ra-224	41	13	27	19			
Ra-226	81	7	10	2			
Ra-228	28	12	25	35			



**Figure 3.** Graph showing reported concentrations, concentrations plus and minus the combined standard uncertainty (CSU), and the minimum detectable concentration (MDC) for Ra-224 (*A*), Ra-226 (*B*), and Ra-228 (*C*), for all samples including duplicates.

highest concentration of all radionuclides in this survey and was from a well completed in a Coastal Plain aquifer in Maryland. Most of the samples (70 percent) contained less than the contractual MDC of 1 pCi/L of Ra-224, with half the concentrations reported less than 0.3 pCi/L (table 4). About 5 percent of the samples exceeded 10 pCi/L (table 5).

The ranked concentrations of Ra-224 were significantly correlated with those of Ra-228 and Ra-226 (table 6). The concentrations of Ra-224 plotted against those of the corresponding Ra-228 concentrations group closely about the 1:1 line (fig. 4A), particularly in the ranges above 1 pCi/L. Comparisons of values less than about 1 pCi/L tend to be at or near the sample-specific MDC and are below the contractual MDC; comparison may be affected by analytical uncertainty; difference in the quality of analytical results obtained at the low concentration levels with the different analytical techniques used, and artifacts. Scatter in relative terms is, therefore, much greater about the 1:1 line for concentration values below 1 pCi/L than above this value. One of the plotted values that deviate substantially from the 1:1 ratio for activity of Ra-224 when compared to Ra-228 when concentrations were above 1 pCi/L include the samples from research wells not used for drinking water in the Coastal Plain Physiographic Province in South Carolina. The Ra-224-rich water samples from South Carolina are highly acidic (pH < 4) and may not be typical of community drinking water supplies throughout the Nation. Ancillary water-quality information was not available for the sample from the State of Missouri that is also an exception to the 1:1 trend for the activity ratios.

The concentration of Ra-224 plotted against those of the corresponding Ra-226 concentrations group do not plot closely about the 1:1 line (fig. 4*B*). Much more scatter around the 1:1 line relative to Ra-226 is apparent than is the case relative to Ra-228 (fig. 4*A* and 4*B*) for concentration ranges both above and below 1 pCi/L; this consistency indicates that the result is not an analytical artifact. Although the ranked concentrations of Ra-224 correlate significantly with those of Ra-226, it is possible to have large quantities of either one and not the other in ground water.

#### Radium-226 and Radium-228

Most of the concentrations of Ra-228 in samples analyzed for the reconnaissance survey are less than the contractual MDC of 1 pCi/L, and half are below 0.5 pCi/L (table 4). The maximum concentration of Ra-228 (72.3 pCi/L) was found in the same Coastal Plain well in Maryland with the maximum Ra-224 concentration. The maximum concentration of Ra-226 was 16.9 pCi/L from a well in Iowa. Most of the concentrations of Ra-226 were less than the contractual MDC of 1 pCi/L, with a median concentration of 0.4 pCi/L.

The Spearman correlation coefficient of Ra-226 and Ra-228 indicates that the ranked concentrations of the two isotopes are significantly correlated (table 6). The general weak nature of co-occurrence of Ra-226 with Ra-228 is illustrated in fig. 4C where it can be seen that many samples do not plot near the 1:1 line.

**Table 4.** Summary of radionuclide concentrations in theUnited States Geological Survey reconnaissance survey

Concentration, in picocuries per liter						
Radionuclide	Mean	Median	Standard deviation	Maximum	Number of samples	
Ra-224	3.2	0.3	10.1	73.6	99	
Ra-226	1.6	0.4	2.8	16.9	99	
Ra-228	2.1	0.5	7.9	72.3	99	
Pb-210	0.6	0.5	0.5	4.1	96	
Po-210	0.1	0.01	0.5	4.9	96	

**Table 5.** Percent of samples reported to exceed specified

 radionuclide concentrations

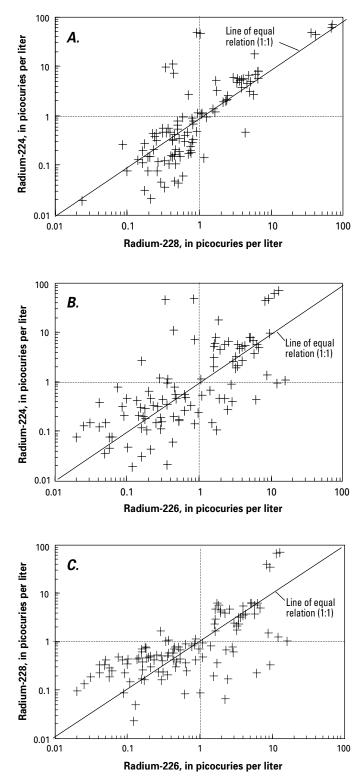
	Total number	Percent of samples that exceed specified concentrations, in picocuries per liter					
Radionuclide		1	2	3	5	7	10
Ra-224	99	30	26	20	15	9	5
Ra-226	99	33	22	17	10	5	2
Ra-228	99	26	18	14	9	2	2
Po-210	95	3	1	1	1	0	0
Pb-210	96	10	3	1	1	0	0

#### **Radium Isotope Ratios and Correlations**

Concentrations of Ra-228 were correlated with those of Ra-224. Radium isotopic ratios may be biased because attempts to account for uncertainty in analytical methodologies was beyond the scope of this report. However, there is a physical basis to the high degree of correlation because both isotopes originate from the same decay series (that of thorium-232), and Ra-224 is indirectly derived from decay of Ra-228 through Th-228. Parsa (1998) reports that the Ra-224:Ra-228 ratios ranged from 1.2 to 2.0 in initial results from the Coastal Plain of New Jersey. The Ra-224:Ra-228 ratios ranged from 1.2 to 2.0 in South Carolina ground water and from 0.8 to 1.7 in Connecticut according to values reported from the literature (Hess and others, 1985). The strong correlations found in the reconnaissance survey corroborate the previous studies and indicate a typical ratio of Ra-224 to Ra-228 of near 1. Thus, the conclusion that Ra-224 and Ra-228 co-occur in nearly equal concentrations is based on many samples collected from a variety of geohydro-

**Table 6.** Spearman correlation coefficient matrix (significant at 0.05 level) for radium isotope concentrations

	Ra-224	Ra-226	Ra-228
Ra-224	1.0	0.69	0.82
Ra-226	_	1.0	0.63
Ra-228		—	1.0



Note: Values of radium isotopes less than 1pCi/L tend to be equal to or near (above or below) the minimum detectable concentration and may not be reasonable for comparison because of the difference in quality of analytical results abtained at low concentrations.

**Figure 4.** Relations of radium-224 to radium-228 (*A*), radium-224 to radium-226 (*B*), and radium-226 to radium-228 (*C*).

logic settings across the Nation. The water samples in this reconnaissance survey were collected from a multitude of aquifers and geochemical environments from most of the major physiographic provinces of the United States. It is thus reasonable for screening purposes to use Ra-228 as a proxy indicator for the much shorter lived and infrequently sampled Ra-224. For example, indications of high concentrations of Ra-228 in water from a particular aquifer or region could help provide an "early warning" to public water-supply systems that analysis for the concentration of Ra-224 and determination of gross alpha-particle activity within 48-to-72 hours after sample collection should be considered. The USEPA suggests that regions suspected of containing Ra-224 in ground water should use the short holding time of 48-to-72 hours for analysis of gross alpha particle activity (USEPA, 2000). Furthermore, a more rigorous analysis of the occurrence and distribution of Ra-228 concentrations in ground water across the Nation could aid in the design of a more complete national survey targeted for Ra-224 than this initial effort was able to provide.

The ranked concentrations of Ra-226 and Ra-228 also correlate significantly (table 6); many samples, however, deviate from the 1:1 ratio (fig. 4*C*). The Ra-228 and Ra-226 concentration data in this study thus are consistent with numerous other studies, which all indicate that Ra-228 and Ra-226 are not uniformly present in a nearly 1:1 ratio in ground water (Hess and others, 1985; Blanchard and others, 1985; Szabo, 1990). Ra-226 is not a good proxy indicator of concentrations of Ra-228 or Ra-224 in ground water, although where Ra-226 is present, Ra-228 and Ra-224 also are likely to be present.

#### Polonium-210 and Lead-210

The maximum concentration of Po-210 was 4.9 pCi/L (table 4) in a sample collected from a well in the Coastal Plain of Virginia. Only one other sample, which had a Po-210 concentration of 1.4 pCi/L, exceeded the contractual MDC of 1 pCi/L. The water chemistry from the well with the highest Po-210 concentration does not provide any information to indicate why Po-210 is likely to be present in elevated concentrations (David Nelms, USGS, Virginia District, written comm. 1999). The radium isotope and Pb-210 concentrations were all less than one (except for the maximum). The pH was 7.5 (near neutral, not acidic), the specific conductance was 259 microsiemens/cm<sup>2</sup> (not unusual for ground water in this Coastal Plain aquifer), the turbidity was 0.53 NTU (below drinking water secondary standards), the alkalinity was 119 mg/L as CaCO<sub>2</sub> (buffered), and there was measurable dissolved oxygen [dissolved oxygen should be 0 in sulfidic waters, the type of waters reported by Harada and others, (1989) to contain Po-210].

The maximum concentration of Pb-210 was 4.1 pCi/L in a sample collected from the Appalachian Province in Connecticut (table 4). Approximately 10 percent of the samples exceeded a Pb-210 concentration of 1 pCi/L (the contractual MDC; table 5), but only one sample exceeded 3 pCi/L. The greatest frequency of detection was for samples from the Appalachian Physiographic Province, especially those from the northeastern United States.

#### LIMITATIONS AND PERSPECTIVE ON THE U.S. GEOLOGICAL SURVEY RECONNAISSANCE SURVEY

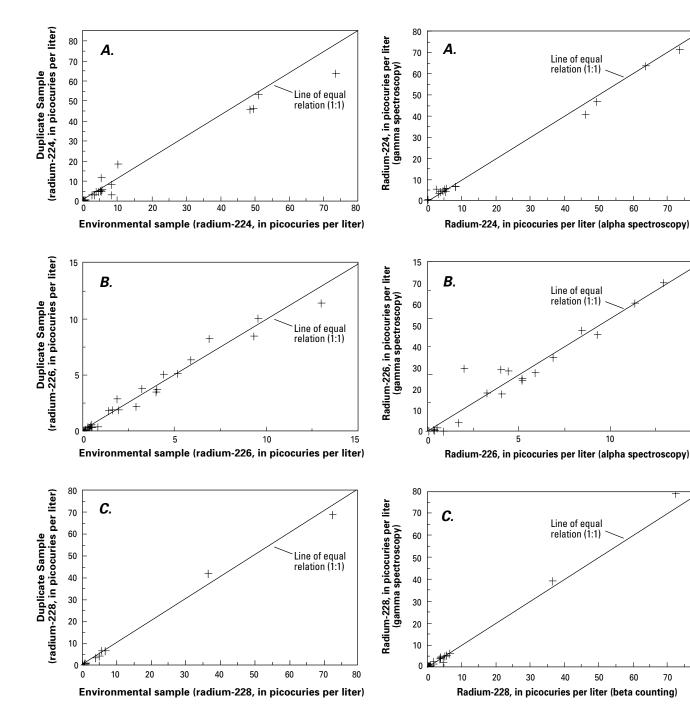
The reconnaissance survey was designed to target areas of known, or suspected, high concentrations of Ra-224 as inferred by associated radium-occurrence data, geologic maps, and other geochemical considerations. Thus, the reconnaissance survey is likely biased toward the high end of the frequency of occurrence distribution for Ra-224 and the co-occurring natural contaminant, Ra-228. Targeted sampling and modeling provides an important evaluation of distribution trends, even in cases where a contaminant is not randomly distributed (Michel and Cothern, 1986). In addition, targeted sampling can be useful in identifying water-quality problems that would be missed or underrated by typical random survey sampling designs and can provide enhanced detail on spatial distributions and variability (Alley, 1993). The results obtained even in this limited targeted sampling thus provide crucial information that could be used to design a comprehensive national analysis of the contributing factors (geologic, hydrologic, geochemical) that control the occurrence distributions of Ra isotopes in various parts of the Nation. The results also fill the data gap in documenting the widespread occurrence of Ra-224 documented to date only in southern New Jersey (Szabo and others, 2000) and add insight to the geologic occurrence of elevated concentrations of Ra-224 in ground water and associated ratios of radium isotopes in various geochemical environments.

#### Analysis of Quality-Control Data

Sequential duplicate samples were collected at about 20 percent of the sampling sites, and preliminary qualitative comparisons were made. The duplicates of the environmental samples analyzed for all radium isotopes with the same techniques indicated reasonable concurrence of results (fig. 5). This result indicates a nearly linear relation among the results of duplicate analyses; the analytical techniques were of suitable quality to provide reproducible results. Although a few pairs of duplicate samples substantially deviate from a 1:1 correspondence for the concentrations of Ra-224 (fig. 5A), the overall fit across the concentration range observed is closely constrained around a 1:1 line. The concentrations of samples analyzed for Ra-224 and Ra-226 by gamma spectrometry are similar to those analyzed by alpha spectrometry (fig. 6A and B). Similarly, the concentrations of Ra-228 analyzed by gamma spectrometry are similar to those analyzed by beta counting (fig. 6C). A more thorough analysis of the quality assurance data is beyond the scope of this report; however, these preliminary results indicate that laboratory methodologies and sample collection are not a significant source of error.

#### The Effects of the Targeted Sampling Design on Interpretations of the Tesults of the U.S. Geological Survey Reconnaissance Survey

The effects on the apparent distribution of radium concentrations due to the targeted sampling design and raw-water sampling can be seen by comparing the results with those of



**Figure 5.** Environmental sample and duplicate environmental sample concentrations for radium-224 (*A*), radium-226 (B), and radium-228 (*C*).

the National Inorganics and Radionuclide Survey (NIRS) (Longtin, 1988) . The NIRS employed a random sampling design stratified by the size (population served) of the individual public water-supply systems and finished-water sampling (although most samples came from systems that were not treating their water). In this way, data collected in this USGS reconnaissance survey from areas of the Nation with known, or presumed, high concentrations of radium in raw ground water are compared with data collected at random

**Figure 6.** Duplicate environmental samples for radium-224 analyzed by alpha spectrometry and gamma spectrometry (*A*), and radium-226 analyzed by alpha spectrometry and gamma spectrometry (*B*), and radium-228 analyzed by beta counting and gamma spectrometry (*C*).

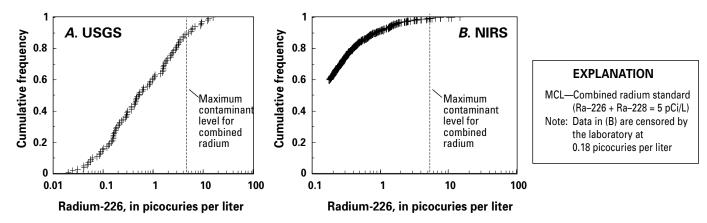
across the Nation from raw and finished ground water (NIRS).

The NIRS data were collected in the public water-supply distribution system and may thus represent effects of the distribution system on water chemistry, as well as effects of water treatment. It is not possible with the data at hand to quantitatively distinguish differences in the USGS reconnaissance and NIRS data due to sampling design and other fac-

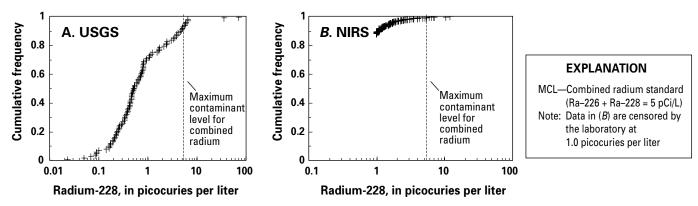
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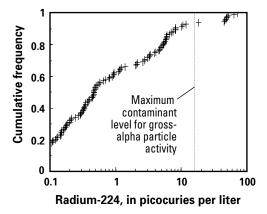
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**Figure 7.** Cumulative frequency distributions for radium-226 in the 1998 U.S. Geological Survey (USGS) reconnaissance survey (*A*; targeted survey design) and in the National Inorganics and Radionuclide Survey (NIRS) (*B*; random survey design). (MCL, maximum contaminatnt level.)



**Figure 8.** Cumulative frequency distributions for radium-228 in the 1998 U.S. Geological Survey (USGS) reconnaissance survey (*A*; targeted survey design) and radium-228 in the National Inorganics and Radionuclide Survey (NIRS) (*B*; random survey design). (MCL, maximum contaminant level.)



**Figure 9.** Cumulative frequency distribution for radium-224 in the 1998 U.S. Geological Survey Reconnaissance survey (targeted sampling design).

tors, such as water treatment and distribution-system effects. Several types of water-treatment practices can remove at least some percentage of radium from water, including lime softening, ion exchange, iron removal, and filtration. A preliminary analysis of the NIRS data revealed that the distribution of concentrations of radium in water samples collected from public water-supply systems with no treatment (the majority of samples in NIRS) were not different than samples collected from systems with treatment (William Labiosa, U.S. Environmental Protection Agency, Office of Ground Water and Drinking Water, written commun., 1999). The treatment technologies associated with the treated water samples in NIRS were not necessarily targeting radium removal. Based on the preliminary analysis, water treatment is not expected to bias comparisons of raw (USGS reconnaissance survey) and finished (NIRS) waters, not because potential treatment techniques are ineffective, but because treatment techniques for many sites surveyed in the NIRS were not used for the purpose of Ra removal. The major differences in interpretations of the distribution of radium concentrations in the two databases are thus most likely due to differences in survey design.

Graphical comparisons of the cumulative frequency distributions of Ra-226 and Ra-228 are illustrated in figs. 7 and 8 respectively (Ra-224 was not analyzed in NIRS). The occurrence of concentrations of Ra-226 above 1, 3, 5, 7, and 10 pCi/ L is more frequent in the targeted sample design of the present USGS reconnaissance survey (fig. 7*A*) than in the random NIRS design (fig. 7*B*). A dashed line indicates the 5 pCi/L concentration level, the level at which the combined radium (Ra-226 + Ra-228) MCL is exceeded. Results from the NIRS random design show that the combined radium MCL is exceeded by Ra-226 alone in less than 1 percent of the samples in the entire United States (fig. 7B). On the other hand, the targeted sampling design (fig. 7A) shows that about 10 percent of the samples exceed the MCL in areas of the country that were presumed to have the types of geology and geochemical environments conducive to uranium and radium in source material and high degrees of radium mobility, thus resulting in high concentrations of Ra-226 in ground water. Comparison of the cumulative frequency distributions of Ra-228 between the random sampling design of NIRS and the targeted sampling design of the USGS reconnaissance survey indicates a similar bias in the data from the targeted sampling. Less than 1 percent exceed 5 pCi/L in the NIRS data (fig. 8B) and about 9 percent exceed 5 pCi/L in the targeted survey data (fig. 8A). The cumulative frequency distribution of Ra-224 from the targeted sampling design (fig. 9) is included for completeness; about 15 percent (table 5) of the samples in the targeted survey exceed a value of 5 pCi/L for Ra-224, and nearly 5 percent of the samples exceed 15 pCi/L, the MCL for gross alpha-particle activity.

The targeted sampling design of the USGS reconnaissance survey clearly results in a data distribution with a greater frequency of concentrations above any given value than in the case of the random sampling (NIRS). For the targeted regions, then, the rate of compliance clearly is lower than for the Nation as a whole and represent the regions where future investigations need to be focused to define the scope of the exposure problem. For the targeted regions, the rate of compliance with the MCL will be a function of the sampling design and will depend on the variability of the Ra distribution within the affected aquifers.

#### Cancer Risk Estimates for Radium Isotopes in Drinking Water

The tables, statistics, and graphics presented in this report summarize the concentrations of the targeted radionuclides. The concentration data, although useful for the many purposes as previously stated, can also be associated with human-health risk information to provide the reader with an additional perspective in interpreting the importance of the results in terms of human-health risk.

In evaluating exposure to radionuclides in drinking water, the USEPA considers only carcinogenic risk for all radionuclides, except uranium (N. Nelson and L. Ralston, U.S. Environmental Protection Agency, Office of Radiation and Indoor Air, written commun., 1999). Uranium is the only element for which chemical toxicity (as a kidney toxin) has been identified to be comparable to or greater than the radiotoxicity, and for which a reference dose has therefore been established to evaluate chemical toxicity. Thus, for radioisotopes of uranium, both chemical toxicity and radiogenic cancer risks are evaluated. Other types of detrimental health effects are also associated with exposure to ionizing radiation, including mutagenic, teratogenic, and acute toxicity effects. These effects are generally less significant for doses associated with environmental exposures. Therefore, the USEPA considers carcinogenic risk to be a sufficient basis for assessing radiation-related human-health risks for drinking water (N. Nelson and L. Ralston, U.S. Environmental Protection Agency, Office of Radiation and Indoor Air, written commun., 1999). For the majority of radionuclides, the risks from any chemical toxicity are much smaller than the radiation-related risk (radiotoxicity), so that only radiological risks normally need to be evaluated (U.S. Environmental Protection Agency, 1999a).

The USEPA estimates of excess fatal (mortality) and total (morbidity) cancer risks for lifetime intakes of Ra-224, Ra-226, and Ra-228 in drinking water (table 7) are obtained from Federal Guidance Report No. 13 (U.S. Environmental Protection Agency, 1999a). The first set of values (columns 2 and 3) in table 7 are the unit concentration risks; that is, the estimated excess fatal and total cancer cases per million per-

Radionuclide Isotope	Estimated number of excess fatal and total cancers per million persons						
	2 liters of drinking	e (70 year) intake of g water per day at: entration† (1 pCi/L)	2 liters of drinking	e (70 year) intake of g water per day at: rent MCL‡			
	Fatal (mortality)	Total (morbidity)	Fatal (mortality)	Total (morbidity)			
Ra-224	5	9	na	na			
Ra-226	14	20	70	100			
Ra-228	38	53	190	265			

 Table 7. U.S. Environmental Protection Agency's cancer risk estimates for radium isotopes in drinking water

 [MCL, maximum contaminant level; na, not applicable; pCi/L, picocuries per liter]

† The risk from 1 pCi/L. From Federal Guidance Report No.13 (U.S. Environmental Protection Agency, 1999), assuming a lifetime intake of 51,100 liters. See text for details.

‡ Assumes that each isotope is ingested alone at the following concentrations in

sons per pCi per liter of water ingested (pCi/L)<sup>-1</sup>, assuming lifetime intake. These values are conversions of USEPA's tap-water mortality and morbidity risk coefficients and USEPA's assumptions for lifetime intake (2 liters of water per day, 365 days per year, for a lifetime of 70 years, equivalent to a total of 51,100 L of water consumed over a lifetime). The unit concentration risk is defined by USEPA (1999a) as:

### $UCR_{F/T} = RC_{F/T} * 0.037 * 51,100$

where

- UCR is the unit concentration risk  $(pCi/L)^{-1}$  for 70-year lifetime for fatal  $(UCR_{E})$  or total  $(UCR_{T})$  cancer cases
- **RC** is the radionuclide-specific tap water risk coefficient in SI units (units of risk per becquerel; Bq<sup>-1</sup>) for fatal  $(RC_{F})$  and total  $(RC_{T})$ ,
- F/T is fatal (mortality) or total (morbidity),

0.037 is a conversion factor (for converting Bq<sup>-1</sup> to pCi<sup>-1</sup>), and 51,100 is the assumed lifetime intake of drinking water (L).

For example, the fatal UCR<sub>r</sub> for Ra-226, given  $RC_{r} = 7.17E-09 Bq^{-1}$  (USEPA, 1999a) is found by converting to the desired units and using USEPA's assumption of lifetime intake: 7.17E-09 x 0.037 x 51,100 = 1.36E-05 or about 14 fatalities per million (table 7). The fatal  $RC_{F}$  for Ra-224 and Ra-228 are 2.74E-09 Bq<sup>-1</sup> and 2.00E-08 Bq<sup>-1</sup>, respectively (USEPA, 1999a). The total  $RC_{T}$  for Ra-224, Ra-226, and Ra-228 are 4.50E-09 Bq<sup>-1</sup>, 1.04E-08 Bq<sup>-1</sup>, and 2.81E-08 Bq<sup>-1</sup>, respectively.

The second set of values (columns 4 and 5) in table 7 are the estimated excess fatal and total cancer cases per million persons assuming lifetime intake of each radionuclide alone at its current associated maximum contaminant level. The risks associated with the current MCLs are:

where

$$MCLR_{F/T} = UCR_{F/T} * MCL$$

\* MCI

MCLR

MCLR is the lifetime excess risk at the MCL for fatal  $(MCLR_{F})$  or total  $(MCLR_{T})$  cancer cases, and

**MCL** is the current maximum contaminant level (pCi/L).

For example, given the UCR<sub> $_{\rm F}$ </sub> of 14 (per 10<sup>6</sup>) for Ra-226 and an MCL of 5 pCi/L, the MCLR<sub>E</sub> for Ra-226 is 70 fatalities per million people.

In this way, the unit concentration risk (USEPA, 1999a) of any of the three radium isotopes listed in table 7 can be used to provide some preliminary perspective on the humanhealth risk associated with consuming 2 L of drinking water every day for a lifetime within the ranges of concentration of radium found in the USGS reconnaissance survey (tables 4 and 5). Each raw-water sample was collected near the wellhead from only one well in each targeted public water-supply system, and each well was sampled only once. Consequently, variability in concentrations of radionuclides due to longterm and seasonal changes over time and other factors are not determined, and an individual sample is representative of the water quality of the local ground-water-flow system contributing water to the targeted well for the time at which it was sampled. The data presented in this report, therefore,

may not be appropriate for estimates of "reasonable maximum exposure" used in risk assessments. This discussion is intended only to provide perspective beyond the presentation of concentration values so that the associated human-health risk of drinking water with the concentrations of radium found in the USGS reconnaissance survey can more easily be understood.

The cancer-risk estimates (table 7) may be interpreted in two ways: either as the age-averaged, lifetime excess probability of cancer induction within a population of individuals of all ages and genders, or as the individual lifetime excess probability of cancer induction for an average or representative member of a given population (N. Nelson and L. Ralston, U.S. Environmental Protection Agency, Office of Radiation and Indoor Air, written commun., 1999). For example, the value of 100 total cancers per million persons, estimated for the lifetime intake of Ra-226 at its current MCL, can also be interpreted as an individual risk of 1 in 10,000 (that is, 100 per  $10^6 = 1$  per  $10^4$ ). Federal Guidance Report No. 13 (USEPA, 1999a) details uncertainties, assumptions, and limitations associated with the cancer-risk estimates.

#### Interpretations of Polonium-210 and Lead-210 Data

Polonium-210 and Pb-210 are derived from the U-238 decay series; the same decay series that produces Ra-226. Because the USGS reconnaissance survey was primarily designed to assess the distribution of Ra-224, some areas in the United States that contain high uranium concentrations in aquifer materials without accompanying high thorium concentrations are likely not represented or are under-represented by the sampling. Such regions may include areas of phosphate rock from Florida or the western United States, or uraniferous sandstones from the Colorado Plateau region. The presence of high concentrations of Po-210 or Pb-210 in ground waters is possible even in the absence of the presence of high concentrations of Ra-224. Results for the dissolved concentration distributions of these radionuclides are possibly biased low, as opposed to the concentration distributions of Ra-224 that are likely biased high. Within these constraints, nevertheless, the Po-210 and Pb-210 concentration data collected as part of the reconnaissance survey can be used to begin to fill the gap in information on the occurrence of these isotopes. These isotopes (Po-210, Pb-210) have recently been proposed for inclusion on the list of contaminants under the Unregulated Contaminant Monitoring Rule (U.S. Environmental Protection Agency, 1999c).

Polonium-210 was found in concentrations exceeding 1 pCi/L in only two wells. The two observations could not be associated with unique geochemical environments as was the case in a previous study from Florida (Harada and others, 1989). With only two results greater than 1 pCi/L, further investigations would be necessary to infer anything more about the national distribution and occurrence of Po-210.

Ten percent of the samples exceeded a Pb-210 concentration of 1 pCi/L; only one sample, however, contained a concentration greater than 3 pCi/L. The greatest frequency of detection was in the Appalachian Physiographic Province of

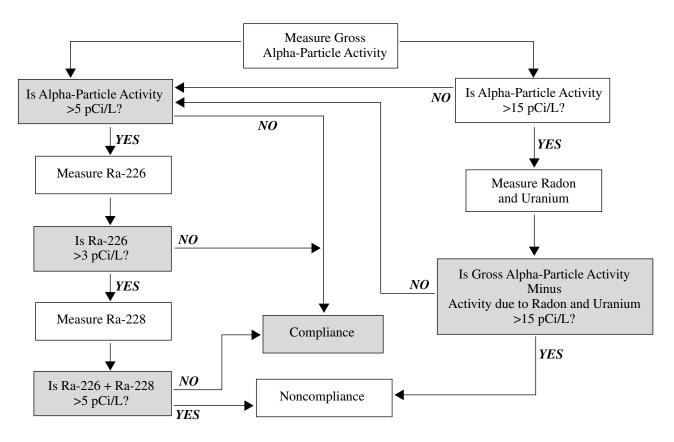
the northeastern United States, especially in the States of Connecticut and Pennsylvania. The geochemical mechanism that controls Pb-210 dissolution is not established and needs further study. It is known that lead is more difficult to extract from sediment material than radium, and adsorbs more quickly onto aquifer solids (Krishnaswami and others, 1982). One possible explanation for the frequent detection of Pb-210 in concentrations greater than 1 pCi/L in the Appalachian region may be the high concentrations of radon-222 in ground water in this region (Zapecza and Szabo, 1987). As the radon in solution decays through a series of very short half-lived products to Pb-210, a small fraction of the Pb-210 may not be immediately sorbed onto the aquifer matrix, or may be only weakly sorbed, or may be re-ejected to the water through alpha recoil after weak adsorption of the radioactive parent product, Po-214. If the latter is true, the higher the initial Rn-222 concentration, the more likely it is that measurable amounts of Pb-210 are detected in the ground water. Regions where Pb-210 concentrations need to be characterized could be identified by elevated Rn-222 and U-238 concentrations in ground water. This hypothesis could not be tested in this initial USGS reconnaissance survey, however, because the Rn-222 concentrations were not analyzed.

#### IMPLICATIONS FOR COMPLIANCE MONITORING

The occurrence of the various radionuclides and the ratios of the three radium isotopes analyzed in the USGS reconnaissance survey indicate that conventional compliance monitoring for gross alpha-particle activities and combined radium may be inadequate in some parts of the United States. This section first describes the commonly used compliance monitoring procedures and then shows their limitations in light of the results of the USGS reconnaissance survey.

#### Compliance Monitoring Procedures for Regulated Radionuclides Used by Public Water-Supply Systems and Results of Previous Studies

Lappenbusch and Cothern (1985) discuss the regulations for radioactivity in drinking water at that time and present a flow chart for compliance monitoring (fig. 10) that is still in conventional use today. The amount of alpha-particle emitting radionuclides are initially determined by analyses of the gross alpha-particle activity of a given water sample. Depending on the gross alpha-particle activity, subsequent analyses may be required. The gross alpha-particle activity screen is viewed as a cost-effective way to determine when additional analyses could be required to quantify the most important alpha-particle



**Figure 10.** Schematic diagram showing the compliance monitoring scheme for radionuclides in publicdrinking-water supplies. Shaded boxes emphasize where this, and other, studies have shown potential for errors in combined radium and short-term gross alpha-particle compliane monitoring. [Modified from Lappenbusch and Cothern (1985).]

emitting radionuclides in a public water-supply system. A combined radium (Ra-226 and Ra-228) standard was associated with this process to avoid the additional costs involved with separate Ra-228 analyses for many public water-supply systems. By use of the gross-alpha particle screening process, a gross alpha-particle activity that does not exceed 5 pCi/L is taken to imply a combined radium concentration that does not exceed 5 pCi/L; similarly, a Ra-226 concentration that does not exceed 3 pCi/L is taken to imply that the Ra-228 concentration does not exceed 2 pCi/L (Blanchard and others, 1985). Hess and others (1985) explain that this scheme, in the absence of actual data, was based on the assumption that Ra-226 was the dominant radioactive component of drinking water and that the Ra-226: Ra-228 ratio was 3:2 or greater.

Data collected from State surveys conducted by the USGS, State Regulators, and university research staff showed a range in the Ra-226: Ra-228 ratio of more than 14 to less than 0.07 (King and others, 1982; Hess and others, 1985; Szabo, 1990; Senior and Vogel, 1995; Kay, 1999). A high degree of correlation between Ra-228 and Ra-226 concentration was not consistently found in most samples, nor were there specific trends in the distribution or ratios of concentrations of these two isotopes by either aquifer, depth, or extent of aquifer when examining large-scale regional or national-scale concentration data (Hess and others, 1985). Blanchard and others (1985) concluded that although the schedule to screen for gross alpha-particle activity has been useful in identifying many initial problem areas, screening has limitations. For example, the screening would miss situations when the gross alpha-particle activity is less than 5 pCi/L, Ra-226 is less than 3 pCi/L, and Ra-228 concentrations are greater than 2 pCi/L; in these instances the presence of elevated concentrations of Ra-228 would go unrecognized. Consequently, where the ratio of Ra-226: Ra-228 is less than 3:2, the gross alpha-particle activity determination is not suitable for combined radium compliance monitoring.

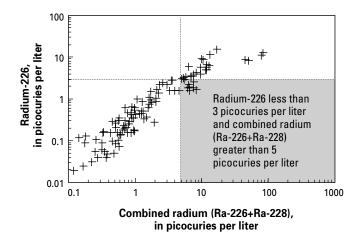
Blanchard and others (1985), even with the limited amount of information available, note that occasionally Th-232, Po-210, or Ra-224 contribute to the alpha-particle activity. The short-lived Ra-224 requires measurements for gross-alpha particle activity to be made within the short time frame before it decays (Parsa, 1998; Szabo and others, 1998a). Regulatory guidelines do not, however, exist to address the limited time required for gross-alpha-particle analyses. Therefore, gross-alpha-particle activity data collected to date for national data bases could be biased low if analyses were not completed before short-lived isotopes decay. Consequently, more public water-supply systems may be out of compliance with the gross alpha-particle activity MCL, and potentially the radium MCL, than has been realized. Szabo and others (1998a) showed that alpha-particle radioactivity in drinking-water supplies from the Kirkwood-Cohansey aquifer system in New Jersey was larger than previously determined in all 12 community water supplies that were re-evaluated with strict adherence to the 48-hour holding time for analysis. The gross alpha-particle activity MCL

of 15 pCi/L was exceeded in all 12 community water supplies when analyzed within 48 hours of collection, whereas for 10 of these wells that were previously sampled for gross alpha-particle activity but analysis was completed at a random time frame after sampling, only 1 was out of compliance. Parsa (1998) outlined the potential effects that short-lived radionuclides such as Ra-224 may have on the monitoring schemes for radionuclides proposed within the Safe Drinking Water Act.

#### Limitations in the Current Monitoring Procedures for Alpha-Particle Emitting Radionuclides and the Combined Radium Standard

The effectiveness of the national drinking water MCL for gross alpha-particle activity and use of this result as a compliance-monitoring screen for combined radium have been shown to be dependent on the holding time between collection and analysis (Parsa, 1998; Szabo and others, 1998a), and the ratios of Ra-226:Ra-228 and Ra-224:Ra 226. The gross alpha-particle activity must be analyzed within about 48-to-72 hours in order to account for the presence of the short-lived alpha-particle-emitting isotopes such as Ra-224. A concurrent study of ground-water supplies by the USGS in cooperation with the State of New Jersey (Szabo and others, 2000) is providing more insight into the role of the distribution of Ra-224 and progeny in determining the distribution of dissolved gross alpha-particle activities. In that study in New Jersey, gross alpha-particle emissions were measured before decay of Ra-224 and after sufficient time had elapsed for nearly complete decay (about 20-to-22 days). The difference between the initial gross-alpha measurement and the final measurement is indicative of the contribution of Ra-224 and all other alpha-particle emitting isotopes that would decay within this time frame. The results indicate that the contribution of Ra-224 and short-lived products is consistent with the theoretical prediction of Parsa (1998) and is approximately three times the concentration of Ra-224. Although this analysis was developed with a small data set in a restricted geographic range, it is based on a physical process. Thus, the result has important implications for USEPA or other regulatory agencies to allow them to project Ra-224 occurrence in association with gross alpha-particle activities and Ra-228 concentrations that are already determined for many public water-supply systems nationwide. The results from New Jersey, allowing for at least an initial estimate of the Ra-224 concentration, are also important in light of both the costliness and difficulty in scheduling the Ra-224 analysis. The results also make clear that in regions where radium is present in ground water, gross alpha-particle activity determined more than 72 hours after sample collection can significantly under represent the actual gross alpha-particle activity of the water.

The gross alpha-particle activity screen, when used to determine compliance with the combined radium (Ra-226 + Ra-228) standard of 5 pCi/L, could produce false negative results for Ra-228. The concentration of Ra-228 is greater than that of Ra-226 in many samples analyzed in this study



**Figure 11.** Graph showing relation between radium-226 and combined radium (radium-226 + radium-228) concentrations.

over the entire range of concentrations of both radium isotopes (fig. 3). The gross alpha-particle activity screen currently in place for combined radium compliance (Hess and others, 1985) relies on Ra-226 concentrations being present in higher concentrations than Ra-228. About 6 percent of the USGS reconnaissance samples contained combined radium concentrations (Ra-226 + Ra-228) greater than 5 pCi/L when Ra-226 was less than 3 pCi/L (fig. 11); these samples would not have been tested for Ra-228 if current traditional compliance monitoring were used (fig. 10).

#### CONCLUSIONS

The USGS reconnaissance survey targeted ground-water supplies of known, or suspected, high concentrations of radium to determine the concentrations of Ra-224 where other isotopes of radium had previously been detected and to determine the co-occurrence characteristics of the three radium isotopes (Ra-224, Ra-226, and Ra-228). About 30, 33, and 26 percent of the samples collected were reported above 1 pCi/L (the contractual minimum detectable concentration) in concentration for Ra-224, Ra-226, and Ra-228, respectively, in the USGS radionuclide reconnaissance survey. About 5 percent of the samples exceeded 10 pCi/L of Ra-224. Several of the samples from the eastern United States (Coastal Plain Physiographic Province) were reported to have pH less than 4.0 and contained detectable concentrations of thorium isotopes, including Th-228, indicating that the analytical result for Ra-224 may be biased high in these areas. Twenty one of the samples exceeded the MCL for combined radium. Six percent of the samples contained combined radium concentrations (Ra-226 and Ra-228) greater than 5 pCi/ L when Ra-226 was less than 3 pCi/L (the ratio of Ra-226:Ra-228 was less than 3:2). Concentrations of Pb-210 were greater than 1 pCi/L in about 10 percent of the samples analyzed; for Po-210, about 3 percent exceeded 1 pCi/L.

Although the number of data points in the USGS reconnaissance survey was limited, it provides new insight into the nationwide occurrence of Ra-224 and the co-occurrence with Ra-226 and Ra-228. The presence of Ra-224 has an effect on compliance with the present drinking-water MCLs for gross

alpha-particle emissions and the associated procedures for compliance monitoring for an unquantified number of community drinking water systems across the Nation. Conventional monitoring procedures, which do not require analysis of gross-alpha-particle activities in time to account for the contribution of short-lived isotopes such as Ra-224, could lead to false indications of compliance. The documented correlation in occurrence and the small variation in the isotopic ratio of Ra-224 and Ra-228 in this study which encompasses several physiographic provinces, indicates that the more frequently monitored Ra-228 is a reasonable proxy to indicate the potential for Ra-224 occurrence. The isotopic ratios of Ra-226 to Ra-228 were below 3:2 in many samples, corroborating previous studies that indicated that the gross-alphaparticle screen commonly used as a first step to determine combined radium (Ra-226 + Ra-228) compliance could be inadequate in some unquantified number of community water systems. Finally, concentrations of Pb-210 and Po-210 were detectable in a small number of well-water samples, although uranium-rich strata were not targeted in the survey.

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### GLOSSARY

**Activity**: The amount of radioactivity in a given volume of such material as water or air. For water, activity is generally expressed in units of picocuries per liter.

Alpha radiation: Alpha radiation is composed of a particle, consisting of two protons and two neutrons, spontaneously emitted from the nucleus of a subset of radioactive elements (mostly the heaviest elements) during radioactive decay. Alpha radiation is ionizing radiation, meaning that it strips electrons from adjacent atoms as it passes. Alpha radiation cannot penetrate skin; thus, an alpha-particle emitting radionuclide must be ingested in order to come into contact with internal tissue. Because of the large size, alpha particles are likely to collide with cell tissue, causing tissue damage. An accumulation of tissue damage in the cell nucleus may lead to cell mutation and potential cancer formation.

Alpha radioactivity, gross: A laboratory measurement of total alpha radioactivity emitted by a sample. This measurement includes radioactivity emitted by Ra-226, Ra-224, Ra-223, Po-210, and numerous other naturally occurring alpha-particle-emitting radioactive isotopes. SHORT-TERM measurement, conducted within 3 days after sample collection, provides information with regard to the amount of alpha radiation present from unsupported, short-lived radionuclides, such as Ra-224. LONG-TERM measurement, usually conducted 20- to-30 days after sample collection, measures only the amount of alpha radiation present from long-lived radionuclides, such as Ra-226 (half-life, 1,602 years).

Alpha spectrometry: An analytical technique that specifies the amount of alpha radiation emitted at specified energy levels, thus allowing determination of individual radionuclide concentrations (from known energy levels of alpha particles unique to each radionuclide). The gridded-pulse-ionization chamber is the most sensitive and most commonly used instrument.

**Beta radioactivity**: Beta radiation is composed of a particle, consisting of an electron, spontaneously emitted from the nucleus of a subset of radioactive elements during radioactive decay. Beta radiation, like alpha radiation, is ionizing radiation—it strips electrons from adjacent atoms as it passes. Beta radiation can only penetrate the surface layer of skin; thus, a beta-particle emitting radionuclide must be ingested in order to come into contact with internal organs or tissues. An accumulation of tissue damage in the cell nucleus may lead to cell mutation and potential cancer formation.

**Beta radioactivity, gross**: A laboratory measurement of total beta radioactivity emitted by a sample. This measurement includes radioactivity emitted by Ra-228 and Pb-210, and numerous other naturally occurring beta-particle-emitting radioactive isotopes. As with gross alpha-particle measurements, both SHORT-TERM and LONG-TERM measurement are possible, providing information regarding the amount of beta radiation present from unsupported short-lived radionuclides and from long-lived radionuclides, such as Ra-228 (half-life, 5.75 yrs).

**Beta radioactivity count**: An analytical technique that specifies total beta radioactivity emitted by a chemically purified sample that contains only one radionuclide of interest. The beta-particle activity is counted in a low-background gas proportional counter. Because detection efficiency is highest for beta particles emitted with the highest energy, the beta-particle activity of a short-lived product of the radionuclide of interest is typically determined. Thus, the concentrations of Ra-228 and Pb-210 are determined by counting the products Ac-228 (actinium-228) and Bi-210 (bismuth-210), respectively.

**Contaminant:** As defined in the Safe Drinking Water Act, any physical, chemical, biological, or radiological substance or matter in water.

**Decay product of radiation**: The isotope remaining after a radioactive decay event.

**Gamma radioactivity**: Gamma radiation is composed of a packet of energy, also known as a photon or photon particle, spontaneously emitted from the nucleus of most radioactive elements during radioactive decay. Gamma radiation is ionizing radiation, meaning that it strips electrons from adjacent atoms as it passes. Gamma radiation can penetrate through skin into internal tissues as opposed to alpha and beta radiation, which cannot. An accumulation of tissue damage in the cell nucleus from gamma radiation may lead to cell mutation and potential cancer formation.

**Gamma spectrometry:** An analytical technique that specifies the amount of gamma radiation emitted at specified energy levels, thus allowing determination of individual radionuclide concentrations (from known energy levels of emitted gamma radiation unique to each radionuclide). A high-efficiency hyper-pure germanium or germanium-lithium detector is typically used for laboratory work requiring low MRLs; the latter was used in the USGS Ra-224 Reconaissance Survey.

**Isotope:** Various forms of a single element differing in the number of neutrons in the nucleus. Unstable isotopes of an element decay through the emission of a form of radioactivity.

Maximum contaminant level (MCL): The maximum permissible level of a contaminant in water that is delivered to the free-flowing outlet of the ultimate user of a public watersupply system (except in the case of turbidity). The MCL for Ra-226, Ra-228, and gross alpha-particle activity in community water systems is:

(a) Combined Ra-226 and Ra-228: 5 pCi/L;

(b) Gross alpha-particle activity (including Ra-226 but excluding radon and uranium): 15 pCi/L.

#### Minimum detectable concentration or activity (MDC):

The minimum quantity of a specific radionuclide detected by the counting instrument for any sample for a given analysis. It is defined by the requirement that the counted radioactivity must differ from the background count by three times the standard deviation of the background count. This value is computed individually for each radionuclide analysis based on instrument operating conditions at the time. A contractual minimum detectable concentration (ASTM, 1999) is contractually determined with the laboratory before the project begins and is generally based on the scope of the project and available resources. The contractual (also called the *a priori*) MDC was targeted for 1 pCi/L for all radionuclides analyzed in this study. For purposes of this paper, the sample-specific (also called *a posteriori*) MDC will be referred to simply as the MDC unless otherwise noted. This value is computed individually for each radionuclide analysis based on instrument operating conditions at the time of measurement and reported by the laboratory for each analysis.

National contaminant occurrence database (NCOD): The Safe Drinking Water Act as amended in 1996 requires

USEPA to develop and maintain a national data base that could be used for regulatory determinations by the Administrator. The first release of the NCOD was in August 1999. The NCOD contains ambient water-quality data obtained by a retrieval from the USGS National Water Information System and data from USEPA's Safe Drinking Water Information System (including compliance data and data collected in anticipation of the Unregulated Contaminant Monitoring Rule). The radionuclide data collected for the USGS Radionuclide Reconnaissance Survey will likely reside in the NCOD. Public access to NCOD is provided by USEPA through the website: www.epa.gov

**Notice of data availablity (NODA):** One of the outlets used by USEPA for public comment and interaction is through a NODA. The NODA for radionclides that USEPA plans to issue will contain all of the existing information that USEPA has on occurrence, health effects, treatment technology, and other pertinent information, including results from the USGS study reported here.

**Negative result:** A result from a radiation measurement (count) of a sample over a fixed interval of time where the measured radioactivity from the sample is less than the longterm average background radiation reported routinely by the instrument.

**Picocurie per liter:** The unit expressing the amount of radioactivity in water. One picocurie per liter equals 2.2 radioactive disintegrations per minute per liter of water.

Radionuclide: An isotope of an element that emits radiation.

**Uncertainty:** A calculated measure of uncertainty of the laboratory analysis. Because radioactive elements decay randomly at any given instant of time, any measurement of radioactivity has an associated uncertainty (also called "counting error") independent of, and in addition to, laboratory sources of analytical uncertainty. Uncertainty can be reported in a variety of ways; the most common include (1) the sum of the laboratory and counting uncertainty, known as "combined standard uncertainty (CSU)" (or the "total propagated uncertainty"), and (2) a counting uncertainty only. There is a 67-percent or 95-percent probability (based on one or two standard deviations, respectively) that the true value of the radionuclide concentration is within the measured value plus

or minus the uncertainty. The uncertainty terms are generally smaller than the measured value except when the measured values are low concentrations (near the MDC). The uncertainties associated with the various radionuclides in this report are (excluding EPA quality control lab):

• One standard deviation of the CSU: Ra-224 and Ra-226 by alpha spectrometry

• Two standard deviation of the CSU: Ra-228 and Pb-210 by beta counting, Po-210 by alpha spectrometry

• One standard deviation of the CSU: Ra-224, Ra-226, and Ra-228 by gamma spectrometry

**Unit concentration risk:** Expression of the health risk of developing or dying from cancer per unit concentration of a harmful substance entering the body (exposure). The current estimate listed in USEPA (Federal Guidance Report No. 13) is that 1 picocurie per liter of Ra-226 is associated with an excess fatal cancer rate of 14 people per million if ingested at a rate of 2 liters per day over a 70-year lifetime. The excess fatal cancers from consuming a unit concentration of Ra-224 is approximately 5 people per million, whereas it is 38 people per million from Ra-228.

## **APPENDIX**

EPA serial number	USGS serial number	Dupli- cate	State	Map key	Latitude (Degrees North)	Longitude (Degrees West)	Region	Geology	Well age (years)	Well depth (feet)	Maximum population served
	44322	1	AK	2	68.85	147.84	2	3	45	85	45000
	44323	2	AK		68.85	147.84	2	3			
	44320	2	AK		68.85	147.85	2	3	16	97	10000
	44321	1	AK	1	68.85	147.85	2	3			
	43372	0	AR	3	36.30	91.52	4	5	26		4200
5914D	45344	0	AZ	4	33.52	111.90	2	1	26	1430	10000
5910Z-1	45345	1	CA	5	34.06	118.08	2	1	75	378	40000
5910Z-2		2	CA		34.06	118.08	2	1			
6040F	45429	0	CA	6	36.59	121.88	2	1	10	481	110000
	44087	1	CA	7	33.94	117.39	2	1	53	415	245000
	44088	2	CA		33.94	117.39	2	1			
	44179	0	CA	8	34.10	117.29	2	1	27	1104	55000
	44319	0	CO	9	40.63	103.22	5	1	14	82	11000
	44328	0	CO	10	40.26	103.62	4	1	6	128	5000
· .	99904	0	СТ	11	41.18	73.19	3	3	33	60	14400
5796R	45219	0	СТ	12	41.00	73.67	3	3	15	52	2340
•	99909	0	СТ	13	41.69	73.23	3	4		560	150
•	99906	0	СТ	14	41.46	72.15	3	4		106	5
· .	99907	0	СТ	15	41.46	72.15	3	4		190	5
5798U	45222	0	СТ	16	41.32	71.90	3	4	8	750	100
•	43553	0	FL	17	27.90	82.78	1	5	38	314	400000
	43468	0	FL	18	28.04	81.96	2	5	16	210	151322
	99908	0	FL	19	27.33	82.54	1	5	5	600	65000
5621T	45066	0	GA	20	32.44	81.77	1	5	22	384	24000
	45426	2	GA	•	33.10	81.99	1	1		•	5700
6038M	45425	1	GA	21	33.10	81.99	1	1			
5620R	45065	0	GA	22	31.49	82.84	1	5	6	705	3000
5659H	45085	0	GA	23	31.60	81.88	1	5	9	890	9000
	45083	2	GA	•	34.77	84.93	3	5		•	21761
5661B	45084	1	GA	24	34.77	84.93	3	5		•	
•	43473	0	IA	25	43.26	91.47	4	2	83	890	4020
•	47128	1	IA	26	41.82	90.63	4	3	4	260	620
•	47129	2	IA	•	41.82	90.63	4	3		•	
•	44325	0	IA	27	40.91	91.39	4	2	38	1850	1900
•	43368	0	IA	28	41.14	91.65	4	2	8	1910	900
•	43475	0	IA	29	42.07	90.83	4	5	15	190	137
•	43407	0	IA	30	42.25	90.43	4	2	51	1500	2400
	43831	1	IA	31	43.32	91.79	4	3	20	70	8063
•	99913	2	IA	· .	43.32	91.79	4	3			
	43951	1	IL	32	41.75	88.29	4	2	40	1719	117000
	43968	2	IL	•	41.75	88.29	4	2			
	44181	2			41.84	88.30	4	2	20	1400	6000
•	44182	1		33	41.84	88.30	4	2			
	43845	2	IL	34	41.71	88.33	4	2	41	1340	4600
	43846	1	IL		41.71	88.33	4	2			
•	44329	1		35	41.84	89.48	4	5	41	1700	6000
60201/	44330	2			41.84	89.48	4	5			
6036K	99914	0		36	40.75	89.60	4	3	59	120	113504
EC71D	44316	0		37	41.76	89.69	4	3	8	130	9700
5671D	45125	0	IN	38	39.74	86.77	4	3	5	134	700
•	44324	0	KS	39	37.97	100.86	5	1	33	377	30000
 	44967	2	KS	40	37.04	100.93	5	1	2	600	16500
5477D	44968	1	KS	•	37.04	100.93	5	1		•	
	45424	2	MD	•	38.99	76.56	1	1		•	450
6025G	45423	1	MD	41	38.99	76.56	1	1	:		<u> </u>
	45422	2	MD		38.99	76.56	1	1	1	100	5
6026H	45421	1	MD	42	38.99	76.56	1	1			· · ·
F011A	99905	0	MD	43	39.15	76.61	1	1	51	115	42000
5911A	45343	0	MD	44	39.53	76.35	3	4	3	500	2000
6299H	45648	0	MI	45	47.24	88.45	4	3	31	100	9228
	43409	0	MI	46	42.73	84.47	4				51000
	43369	0	MI	47	42.70	84.55	4	2	28	330	100000
•	43408	0	MI	48	42.46	84.17	4				1200
	43544	0	MN	49	45.08	93.25	4	2	38	840	29000
5547A	99911	1	MN	50	44.45	95.78	4	3	.		12000
5547A		2	MN	· .	44.45	95.78	4	3	· ·		· · ·
•	43370	0	MN	51	43.62	95.60	4	3	10	106	10000
	44177	1	MN	52	46.01	92.89	4	2	1	390	1200

EPA serial number	USGS serial number	Dupli- cate	State	Map key	Latitude (Degrees North)	Longitude (Degrees West)	Region	Geology	Well age (years)	Well depth (feet)	Maximum population served
	44178	2	MN	-	46.01	92.89	4	2			
	99912	2	MN		46.01	92.89	4	2			
	99910	0	MN	53	44.45	93.16	4	2	21	388	10500
6160N	45603	0	MO	54	39.16	91.87	4	5	14	1500	14000
	43949	2	MO		37.55	94.51	4	5	25	825	690
	43950	1	MO	55	37.55	94.51	4	5		-	•
	43833	0	MO	56	37.71	91.12	4	5	20	680	748
	43546	0	MO	57	37.16	94.31	4	2	7	1500	11000
	43552	0	MO	58	37.14	94.47	4	5	86	1015	1875
6037L	45428	0	MO	59	38.76	93.72	4	2			7500
•	43824	0	MO	60	36.96	93.72	4	5	10	1300	6500
•	43467	0	MO	61	37.60	93.41	4	5	46	1055	8370
•	47127	0	MO	62	37.92	90.54	4	5	10	700	4000
•	43472	0	MO	63 64	37.84	94.35	4	5	14	1200	10000
•	99903			-	35.17	78.98	15		18 4	90 405	8000
•	44084		NE	65 66	42.11	102.88	5		4 2	405	9800
	44180 44085	2	NE NE	00	42.67 42.82	103.40	5		2 29	102 265	1200 6000
	44085		NE	67	42.82	103.00	5		29	200	6000
5662C-1	44086 45087		NJ	68	39.44	75.22	1		33	120	18000
5662C-2	45087	2	NJ	00	39.44	75.22		1		120	18000
5660A-1	45088		NJ	69	39.44	75.00			65	158	32000
5660A-2	45089	2	NJ	09	39.48	75.00		1	05	100	32000
3000A 2	45050	1	NJ	70	40.34	74.44		1	45	120	5605
	46850	2	NJ	/0	40.34	74.44		1		120	3003
	43554		NJ	71	39.92	74.20	1	1	2	280	8000
	43622	2	NJ	''	39.81	74.20			9	248	1950
	43623		NJ	72	39.81	74.24		1	5	240	1550
5800U	45224	0	NJ	73	40.71	75.03	3	2			4977
5797T	45223	0	NJ	74	40.71	74.36	3	2	-	•	19757
5924F-1	45352	1	NM	75	34.41	103.22	5	1	60	368	30000
5924F-2	10002	2	NM	10	34.41	103.22	5	1	00	000	00000
5478E-1	44966	1	ОК	76	36.69	101.47	5	1	33	360	7800
5478E-2		2	ОК		36.69	101.47	5	1			
	43832	Ō	PA	77	40.20	75.09	3	2		-	35000
	44176	0	PA	78	41.05	76.24	3	5	39	87	16000
	44239	0	PA	79	40.78	76.23	3	2			4700
5479F-A	44969	1	sc	80	33.32	81.43	1	1			0
5479F-B	44970	2	sc		33.32	81.43	1	1			
5490A-A	44971	2	sc		33.32	81.43	1	1			0
5490A-B	44972	2	SC		33.32	81.43	1	1			
5799V	45220	1	SC	81	33.12	81.31	1	1			0
	45221	2	SC		33.12	81.31	1	1			
	99902	0	SC	82	33.73	81.10	1	1			500
5517U	45002	0	SC	83	34.28	81.61	3	4	25	110	600
	45052	2	SC		34.21	81.53	3	4	21	465	600
5546Z	45051	1	SC	84	34.21	81.53	3	4		-	
.	43948	1	SD	85	43.43	103.48	5	1	47	26	1800
5663D	45086	2	SD	.	43.43	103.48	5	1	-	21	2500
.	44175	0	SD	86	43.37	99.85	5	1	17	230	5000
	99901	0	ТХ	87	29.70	95.46	2	1	29	1630	2209174
5912B	45348	1	ТХ	88	29.78	95.18	1	1	17	1020	3500
	45349	2	ТХ	.	29.78	95.18	1	1			
5913C	45351	1	ТХ	89	29.78	95.18	1	1	24	1242	2850
.	45350	2	TX		29.78	95.18	1	1			
	43411	2	ТХ		30.74	99.22	5	1	27	437	2043
.	44080	1	ТХ	90	30.74	99.22	5	1			2043
.	44078	0	ТХ	91	30.74	99.22	5	1	15	340	2043
.	43428	2	ТХ	92	30.74	99.22	5	1			2000
.	44079	1	ТХ	.	30.74	99.22	5	1	2	355	2043
	43477	1	ТХ	93	31.16	98.73	5	5	3	120	2626
	43493	2	ТХ		31.16	98.73	5	5			
.	43969	0	VA	94	37.77	77.32	1	1	5	487	400
.	43914	1	VA	95	36.93	77.44	1	1	10	88	100
.	43915	2	VA	.	36.93	77.44	1	1			
.	43410	0	WI	96	43.94	88.09	4	5	75	981	3500
	43371	0	WI	97	43.48	89.74	4	2	37	387	11000
	43545	0	WI	98	43.29	89.74	4	2			2380
	43474	0	WI	99	43.26	89.74	4	2	7	565	1430

USGS	Мар				Ci/L) - USGS			_		24 (pCi/L) - E	· · · ·		
serial number	key	alp result	ha spectro CSU <sup>1</sup>	scopy MDC	gan result	nma spectro: CSU <sup>1</sup>	scopy na	result	alpha spec CSU <sup>2</sup>	troscopy MDC	ga ga	mma spec CSU <sup>2</sup>	troscopy MDC
44322	2	-1.20E-02	1.40E-01	2.50E-01									
44323	-	.72E-02	1.50E-01	2.40E-01									
44320		-2.00E-01	2.80E-01	5.10E-01									
44321	1	3.20E-02	1.86E-01	3.20E-01							.		
43372	3	3.64E-02	1.30E-01	2.30E-01			•	· ·	•			•	•
45344	4	3.30E-01	3.20E-01	4.90E-01			•	2.05E-01	1.50E-01	3.20E-02	4.10E-01	5.50E-01	2.80E-01
45345	5	2.00E-03	1.78E-01	3.20E-01	•	•	•	1.70E-01	1.70E-01	4.30E-02	3.93E-01	6.10E-01	3.00E-01
•	•	•	•	•	•	•	•	6.90E-02	9.80E-02	2.80E-02	·	•	•
45429	6	9.30E-01	3.20E-01	3.50E-01	•	•	•	1.67E-01	1.30E-01	3.20E-02	8.81E-01	7.10E-01	3.00E-01
44087	7	1.90E-01	2.20E-01	3.70E-01	3.80E-01	8.00E-02		•	•	•	·	•	•
44088	•	3.34E-01	1.86E-01	2.60E-01	•	•	•	•	•	•	· ·	•	•
44179	8	1.50E-01	2.40E-01	3.90E-01	•	•	•	· ·	•	•	·	•	•
44319	9	2.80E+00	2.00E+00	3.10E+00	•	•	•	· ·	•	•	· ·	•	•
44328	10	4.84E-01	1.94E-01	2.40E-01	· ·	1 COT 01	•	· ·	•	•	· ·	•	•
99904 45219	11 12	2.00E-02	2.00E-01	3.70E-01	8.00E-02	1.60E-01		1.52E-01	1.50E-01	3.90E-02			1 105.00
		2.00E-02	2.00E-01	3.70E-01	1 445.00	1.50E-01	•	1.52E-01	1.50E-01	3.90E-02	:		1.10E+00
99909 99906	13 14				1.44E+00 -1.60E-01	1.90E-01					:		
99907	14	-	-		-7.00E-01	7.00E-01			_			-	
45222	16	1.23E-01	1.94E-01	3.20E-01	-7.00L-02	7.00L-02		8 60E-02	1.30E-01	3.70E-02		-	9.20E-01
43553	17	1.50E-01	5.00E-01	8.40E-01				0.002-02	1.002-01	J.70L-0Z	.		J.ZUL-UI
43468	18	-5.55E-02	2.80E-01	5.10E-01									
99908	19	5.55E 02	2.001 01	3.10E 01	4.60E-01	1.20E-01							
45066	20	2.70E-01	4.00E-01	6.70E-01	-	.202 01		-5.36F-03	7.60E-02	3.10E-02	5.00E-01	5.80E-01	3.00E-01
45426	-	.53E-01	1.88E-01	2.50E-01								•	•
45425	21	4.70E-01	2.60E-01	3.20E-01	6.60E-01	1.10E-01		3.95E-01	1.70E-01	2.60E-02	8.18E-01	6.50E-01	3.50E-01
45065	22	2.90E-01	2.20E-01	3.10E-01					1.80E-01	3.30E-02	5.67E-01	6.60E-01	3.20E-01
45085	23	1.13E-01	1.58E-01	2.60E-01		•	•		9.00E-02	3.00E-02		•	1.20E+00
45083	•	-1.80E-01	2.40E-01	4.60E-01		•	•		•	•	· ·	•	•
45084	24	4.70E-02	1.52E-01	2.70E-01	•	•	•	4.30E-02	1.10E-01	3.40E-02	· ·	•	8.30E-01
43473	25	1.09E-01	1.34E-01	2.00E-01	:	:	•		•	•	· ·	•	•
47128	26	2.20E-02	9.40E-02	1.60E-01	2.10E-	01 9.50E-0	)2.	· ·	•	•	· ·	•	•
47129	•	2.80E-02	8.40E-02	1.40E-01	•	•	•	· ·	•	•	· ·	•	•
44325	27	1.11E+00	6.60E-01	8.40E-01	•	•	•	•	•	•	· ·	•	•
43368	28	9.64E-01	8.20E-01	1.20E+0	•	•	•	· ·	•	•	· ·	•	•
43475	29	1.52E-01	5.80E-01	1.00E+00	•	•	•	•	•	•	· ·	•	•
43407	30	2.15E+00	6.20E-01	4.60E-01	•	•	•	· ·	•	•	· ·	•	•
43831	31	2.02E-01	1.46E-01	2.20E-01	•	•	•	•	-	•	· ·	•	•
99913	•	•	• • • • • •	•	•	•	•	•	•	•	· ·	•	•
43951	32	8.30E+00	1.28E+00	4.50E-01	6.44E+00	5.90E-01		· ·	•	•	· ·	•	•
43968	•	8.20E+00	1.26E+00	3.60E-01	6.76E+00	5.10E-01		· ·	•	•	· ·	•	•
44181	•	5.54E+00	9.00E-01	4.90E-01		•	•	•	•	•	· ·	•	•
44182	33	5.13E+00	7.60E-01	3.60E-01	5.43E+00	4.10E-01		· ·	•	•	· ·	•	•
43845	34	4.76E+00	9.40E-01	6.80E-01			•	•	-	•		-	•
43846	• २८	3.96E+00		6.00E-01	4.38E+00	4.00E-01							
44329 44330	35	5.34E+00 4.93E+00	7.60E-01 6.80E-01	3.50E-01 2.70E-01	4.36E+00	3.40E-01	-	1					
44330 99914	36	4.JOE+UU	6.80E-01	2.70E-01	:			8 ຍາະ ກາ	9.00E-02	1 805.02			8.50E-01
44316	30 37	5.80E-01	6.00E-01	9.00E-01				0.00E-02	3.00E-0Z	1.00E-02	.		0.300-01
44310	38	3.50E-01	2.40E-01	3.40E-01				3.34F-01	1.80E-01	2 80F-02	6.17E-01	6 10F-01	2.50E-01
44324	39	4.40E-01	2.40E-01 2.20E-01	3.10E-01				+L-01	1.00⊑-01	2.002-02		J. TOL-UT	2.300-01
44967	40	4.90E-01	2.20E-01 2.80E-01	3.90E-01									
44968	•	4.49E-01	1.60E-01	2.00E-01	•			2.33E-01	1.20E-01	2.20F-02	7.46F-01	4.60E-01	2.30F-01
45424		4.62E+01		8.30E-01	4.09E+01	3.52E+00							-
45423	41	4.96E+01		1.10E+00	4.70E+01	3.49E-01		4.56F+01	4.20E+00	2.00E-02	4.82F+01	3.10E+00	1.50F+00
45422	•	6.37E+01		9.70E-01	6.36E+01	4.73E-01							
45421	42	7.36E+01		1.00E+00	7.13E+01	5.40E+00		6.61E+01	5.40E+00	2.70E-02	7.54E+01	4.60E+00	2.40E+00
99905	43	•	•	•	6.57E+00	5.70E-01			•			•	
45343	44	8.00E-02	2.20E-01	3.90E-01	•	•	•	-3.36E-02	1.60E-01	6.50E-02	5.57E-01	6.80E-01	3.30E-01
45648	45	1.31E-01	1.16E-01	1.80E-01	•	•	•	1.60E-02	8.80E-02	3.10E-02	·	•	9.60E-01
43409	46	2.02E+00		4.60E-01	· ·	•	•	· ·	•	•	·	•	•
43369	47	2.62E+00	7.00E-01	6.30E-01	•	•	•	•		•	· ·	•	
43408	48		4.20E-01	5.50E-01	· ·	•	•	· ·	•	•	·	•	•
43544	49	7.05E+00	1.16E+00	4.70E-01	•	•	•	· ·	•	•	·	•	•
	50	•	•	•	6.60E-01	8.00E-02		3.74E-01	3.30E-01	8.10E-02	1	1.20E+00	
99911	50												
99911 - 43370	51	1.17E+00	· ·	3.30E-01	:	•			:		1.32E+00	9.60E-01	4.90E-01

USGS				Ra224 (p0	Ci/L) - USGS				Ra2	24 (pCi/L) - E	PA (quality-o	control lab)	
serial	Map key		ha spectro			ima spectro	.,		alpha spec			mma spec	
number		result	CSU <sup>1</sup>	MDC	result	CSU <sup>1</sup>	na	result	CSU <sup>2</sup>	MDC	result	CSU <sup>2</sup>	MDC
44177	52	2.79E+00	5.20E-01	5.60E-01	5.36E+00	4.20E-01		· ·	•	•	•	•	•
44178 99912	:	3.34E+00	5.80E-01	3.50E-01	5.90E+00	• 4.50E-01	•	:	:	:			:
99910	53				1.19E+00	1.00E-01		.					
45603	54	4.70E-01	3.00E-01	4.20E-01	•	•	•	6.11E-01	3.30E-01	5.50E-02	7.06E-01	7.00E-01	4.50E+00
43949	•	3.15E-01	2.20E-01	2.80E-01	•	•	•	·	•	•	•	•	•
43950	55	4.06E-01	3.00E-01	4.00E-01	:	:	:	:	:	:		:	:
43833 43546	56 57	7.61E+00 5.43E-01	1.66E+00 4.80E-01	1.30E+00 6.80E-01				.					
43552	58	5.80E-01	3.00E-01	3.70E-01	-			.					
45428	59	1.10E-01	4.40E-01	7.70E-01	-	•		7.67E-01	3.10E-01	5.10E-02	5.27E-01	7.60E-01	4.00E-01
43824	60	-2.00E-02	2.80E-01	5.20E-01	•	•	-	·	•	•	•	•	•
43467	61	1.79E-01	1.60E-01	2.30E-01							•		•
47127 43472	62 63	6.04E+00 1.41E+00	1.28E+00 5.40E-01	1.10E+00 6.30E-01		:	:	:	:				:
99903	64	•	J.40L 01	0.002 01	5.95E+00	4.70E-01		.		•	•	•	
44084	65	8.00E-02	3.00E-01	5.00E-01	•	•	•	•	•	•	•	•	•
44180	66	4.50E-02	1.54E-01	2.60E-01	•	•	•	· ·	•	•	•	•	•
44085		4.60E-02	1.72E-01	3.00E-01	· 2.40⊑.01	- 7 00 - 02	•						:
44086 45087	67 68	1.28E-01 5.72E+00	1.48E-01 1.00E+00	2.40E-01 5.50E-01	3.40E-01 5.91E+00	7.00E-02 4.90E-01		5.89E+00	8.10E-01	2.40E-02	7.08E+00	9.20E-01	4.40E-01
45088	•	5.93E+00	9.00E-01	5.80E-01	•	•			•			.201	•
45089	69	3.39E+00	6.20E-01	4.70E-01	3.52E+00	3.40E-01		3.20E+00	5.00E-01	2.50E-02	3.72E+00	8.60E-01	4.10E-01
45090	•	3.03E+00	8.80E-01	7.90E-01	•	•	•	· ·	•	•	•	•	•
46849 46850	70	4.58E+00 4.95E+00	8.00E-01 9.00E-01	3.90E-01 4.20E-01	4.57E+00	3.75E-01		:	:	:		:	÷
40050	71	4.95E+00 1.83E-01	9.00E-01 1.16E+00	4.20E-01 2.90E-01				.					
43622		3.53E-01	2.80E-01	4.00E-01	•			.		•	•	•	•
43623	72	3.07E-01	1.58E-01	2.10E-01	•	•	•	· ·	•	•	•	•	•
45224	73	1.23E+00	5.20E-01	6.10E-01	•	•	•	6.30E-01	2.50E-01	3.20E-02	1.08E+00	6.30E-01	3.10E-01
45223 45352	74 75	3.00E-01 5.00E-02	2.20E-01 1.94E-01	3.30E-01				2.03E-01	1.50E-01 9.00E-02	3.30E-02	9.26E-01	7.60E-01	3.70E-01 2.30E+00
40302	. 1	0.00E-02	1.940-01	3.40E-01				0.00E+00	9.00E-02	3.00E-02			2.30E+00 1.60E+00
44966	76	2.15E-01	1.72E-01	2.70E-01	•		•	1.04E-01	1.60E-01	4.70E-02	•	•	1.00E+00
•	•		•	•	•	•	•	1.57E-01	1.10E-01	2.20E-02	•	•	•
43832	77	1.63E-01	9.60E-02	1.40E-01	•	•			•	•	•	•	•
44176 44239	78 79	3.38E-01 1.50E-01	1.56E-01 2.20E-01	2.10E-01 3.70E-01				:					:
44969	80	5.10E+01	8.40E+00	4.10E+00	3.33E+01	2.50E+00		2.98F+01	5.30E+00	1.30F-01	2.07F+01	2.00E+00	9.90E-01
44970	•	5.32E+01	9.80E+00	2.30E+00	•	•	•		•			1.80E+00	
44971	•	4.86E+01	8.00E+00	2.80E+00	1.65E+01	1.20E-00		3.09E+01	7.80E+00	2.60E-01		2.00E+00	
44972	•	4.58E+01	8.00E+00	3.80E+00	• 6.38E+00	E 00E 01	•	E 16E.00	• 0 40E 01	- 		1.80E+00	
45220 45221	81	5.50E+00 1.18E+01	4.40E+00 2.60E+00	6.80E+00 1.60E+00	0.30E+00	5.00E-01		5.16E+00	9.40E-01	5.80E-02	7.40E+00	8.60E-01	4.20E-01
99902	82		-	1.002+00	3.90E-01	8.00E-02		.					
45002	83	-6.40E-02	1.96E-01	3.60E-01	•	•	•	1.56E-01	1.30E-01	3.00E-02	•	•	7.80E-01
45052	•	2.03E-01	1.20E-01	1.70E-01	•	•	•	· · · ·	•	•	•	•	•
45051	84 85	1.65E-01	1.12E-01	1.70E-01 4.10E+00		:	:	2.24E-01	1.70E-01	3.70E-02		:	9.10E-01
43948 45086	•	9.00E-01 8.20E-01	2.40E+00 1.00E+00	4.10E+00 1.60E+00	•			2,15F-01	1.50E-01	3.20F-02	•		7.10E-01
44175	86	2.47E-01	1.94E-01	2.60E-01	•	•	•				•	-	
99901	87		•	•	8.00E-01	8.00E-02			•	•	•	•	•
45348	88	5.13E-01	1.96E-01	2.30E-01		:	:	4.72E-01	1.40E-01	1.80E-02	8.29E-01	7.50E-01	3.70E-01
45349 45351	89	7.10E-01 4.70E-01	2.80E-01 2.00E-01	3.20E-01 2.90E-01	7.40E-01	2.20E-01		2.92E-01	2.40E-01	5.10E-02	1.34F±00	- 8.80E-01	4.40E-01
45350	•	4.85E-01	1.94E-01	2.30E-01 2.30E-01	· ·	2.20L-01		·	2.70L-01 •	J. TOL-UZ	·	J.UUL-UI	-T.TUL-UI
43411	•	3.25E+00	7.00E-01	2.50E-01	•	•	•	·	•	•	•	•	•
44080	90	8.31E+00	1.50E+00	5.60E-01	•	•	•	· ·	•	•	•	•	•
44078	91	1.06E+01	1.76E+00	6.50E-01		:	:	:	:				:
43428 44079	92	1.85E+01 1.02E+01	2.40E+00 1.68E+00	3.40E-01 5.00E-01				.					
43477	93	3.80E-01	2.80E-01	4.20E-01	•	•	•	.		•	•	•	•
43493	•	2.10E-01	3.00E-01	4.60E-01	•	•	•	·	•	•	•	•	•
43969	94	6.08E-02	1.26E-01	2.10E-01	•	•	•		•	•	•	•	•
43914	95	3.22E-01	1.90E-01	2.40E-01		:	:		:	:		:	:
43915 43410	96	4.69E-01 1.46E-01	2.00E-01 1.90E-01	2.50E-01 3.10E-01				.					
43371	97	2.12E+00	4.60E-01	3.30E-01	•		•	· ·	•	•	•	-	•
43545	98	6.11E+00	1.32E+00	6.30E-01	•	•	•	·	•	•	•	•	•
43474	99	4.11E+00	6.20E-01	3.20E-01	•	•	•	•	•	•	•	•	•

USGS				Ra226 (p	Ci/L) - USG	S			Ra226	6 (pCi/L) - E	PA (quality-	control lab	)
serial	Map key		oha spectro		-	nma spectros			pha spectro		-	nma spectr	
number	-	result	CSU <sup>1</sup>	MDC	result	CSU <sup>1</sup>	na	result	CSU <sup>2</sup>	MDC	result	CSU <sup>2</sup>	MDC
44322	2	1.30E-01	4.00E-02	3.40E-02	•	•	•		•	•	•	•	•
44323	•	1.31E-01	4.20E-02	3.90E-02			•				-	•	
44320	:	1.09E-01	5.20E-02	6.80E-02			•					-	
44321	1	1.59E-01	5.20E-02	5.10E-02			•		•			-	
43372	3	4.92E-02	3.80E-02	5.10E-02	•	•	•	1 00 01	7 405 00		4 105 01		0.00F 01
45344	4	8.60E-02	1.00E-01	1.50E-01	•	•	•	1.02E-01	7.40E-02	1.80E-02	4.18E-01	2.40E-01	2.80E-01
45345	5	1.64E-01	6.00E-02	5.30E-02	•	•	•	1.40E-01	1.00E-01	2.50E-02	•	•	7.40E-01
45429	6	2.87E+00	4.80E-01	7.90E-02			•	9.20E-02 1.22E+00	7.40E-02 2.50E-01	2.10E-02	1.67E+00	3.40E-01	4.30E-01
43429	0 7	2.87E+00 3.00E-01	4.60E-01 9.00E-02	7.60E-02	2.00E-01	3.00E-02	•	1.22E+00	2.30E-01	1.50E-02	1.07E+00	3.40E-01	4.30E-01
44087		2.72E-01	9.00E-02 7.80E-02	4.80E-02	2.00E-01	3.00E-02		· ·	•			-	•
44179	8	2.55E-01	1.28E-01	4.00E-02 1.20E-01	•	•	•		•	•	•	•	•
44319	9	1.60E-01	4.00E-01	7.10E-01		•	•	· ·	•		•	•	•
44328	10	2.13E-01	6.80E-02	5.70E-02		•	•	· ·	•		•		•
99904	11	2.102 01	0.002 02	0.702 02	9.00E-02	3.00E-02	•		•				•
45219	12	1.21E-01	4.40E-02	3.90E-02	0.002 02	0.002 02		3.07E-01	1.40E-01	2.20E-02	5.25E-01	3.20E-01	6.90E-01
99909	13		1.102 02	0.002 02	2.84E+00	2.90E-01	•	0.072 01	1.102 01	2.202 02	0.202 01	0.202 01	0.002 01
99906	14				1.00E+00	1.00E-01							
99907	15				2.27E+00	1.70E-01							
45222	16	1.61E-01	5.20E-01	3.80E-02				2.27E-01	1.10E-01	1.60E-02	6.47E-01	2.90E-01	6.50E-01
43553	17	1.54E+00	3.20E-01	1.20E-01				.					
43468	18	5.17E-01	1.40E-01	1.10E-01									
99908	19		-		6.21E+00	4.60E-01							
45066	20	6.30E-01	2.20E-01	1.70E-01				5.72E-01	1.70E-01	1.70E-02	3.95E-01	3.20E-01	6.40E-01
45426		3.61E-01	9.80E-02	5.50E-02									
45425	21	4.85E-01	1.40E-01	7.70E-02	3.70E-01	5.00E-02		3.11E-01	1.30E-01	1.70E-02			6.90E-01
45065	22	2.49E+00	3.60E-01	7.30E-02				2.73E+00	4.40E-01	1.80E-02	1.97E+00	3.50E-01	4.20E-01
45085	23	2.93E-01	8.00E-02	5.60E-02				3.28E-01	1.40E-01	1.60E-02			6.60E-01
45083		1.09E-01	6.20E-02	7.20E-02									
45084	24	5.70E-02	4.40E-02	6.30E-02				1.38E-01	8.20E-02	1.60E-02	-		6.60E-01
43473	25	1.88E-01	5.40E-02	3.30E-02							-		
47128	26	3.66E-01	6.80E-02	2.40E-02	6.20E-	01 6.00E-02					-	•	
47129		3.23E-01	6.00E-02	2.10E-02									
44325	27	1.62E+01	2.40E+00	1.90E-01		•	•					•	
43368	28	1.23E+01	1.98E+00	1.80E-01		•	•					•	
43475	29	3.13E-02	1.12E-01	2.20E-01	•	•	•		•		-	•	•
43407	30	3.48E+00	6.40E-01	8.90E-02			•					-	
43831	31	4.48E-01	9.60E-02	6.10E-02			•		•		-	-	
99913		F 10F .00	7 005 01		4 505 .00	0.505.01	•	•	•	•	•	•	•
43951	32	5.16E+00	7.60E-01	8.10E-02	4.59E+00	3.50E-01			•	•	•	•	•
43968	•	5.18E+00	7.60E-01	8.10E-02	4.70E+00	3.80E-01			•	•	•	•	•
44181		8.22E+00 6.90E+00	1.18E+00	4.50E-02	C C C O O O	E 20E 01	•		•	•	•	•	•
44182	33		9.20E-01	4.20E-02 5.50E-02	6.62E+00	5.20E-01		· ·	•	•	-	-	•
43845 43846	34	6.39E+00 5.88E+00	9.80E-01 8.80E-01	5.50E-02 6.00E-02	5.23E+00	4.30E-01	•	· ·	•	•	-	-	•
43846	35		5.20E-01	4.70E-02	3.34E+00	2.80E-01		· ·	•			-	•
44329 44330	30	4.06E+00 3.72E+00	5.20E-01 4.80E-01	4.70E-02 3.60E-02	J.J4E+UU	2.00E-01		· ·	•	·	•	•	•
99914	36	J.72LTUU	-T.UUL-UI	0.00L-0Z	•	•	•	3 50F-02	4.00E-02	1 20F-02	•	•	7.00E-01
44316	30	4.70E-01	2.80E-01	2.60E-01	•	•	•	0.002-02	4.00L-0Z	1.202-02	· ·	•	7.002-01
45125	38	7.86E-01	1.56E-01	4.30E-01	•	•	•	7.28E-01	1.80E-01	1.50E-02	5.96E-01	3.00E-01	3.60E-01
44324	39	1.37E-01	6.00E-01	7.10E-02	•	•	•	1.200-01			J.JOL-01	3.00L-01	0.001 01
44967	40	4.08E-01	1.26E-01	1.20E-01	•						· ·	•	•
44968		3.16E-01	7.80E-02	6.00E-02				2.82E-01	1.00E-01	1.20E-02	1.58E+00	3.30E-01	4.00E-01
45424		8.48E+00	1.20E+00	7.80E-02	9.04E+00	7.30E-01	-						
45423	41	9.31E+00	1.46E+00	8.50E-02	8.63E+00	6.70E-01		7.64E+00	8.60E-01	1.30E-02	6.89E+00	5.90E-01	6.90E-01
45422		1.14E+01	1.66E+00	4.70E-02	1.14E+01	8.90E-01						= • •	
45421	42	1.30E+01	1.92E+00	4.30E-02	1.32E+01	1.00E+00		1.19E+01	1.10E+00	1.30E-02	1.06E+01	7.20E-01	7.80E-01
99905	43				2.45E+00	1.90E-01							
45343	44	5.70E-02	5.60E-02					1.71E-01	1.30E-01	3.40E-02	3.84E-01	3.40E-01	5.20E-01
45648	45	2.50E-02	2.60E-02	3.90E-02				0.00E+00	3.60E-02	2.20E-02			7.60E-01
43409	46	3.29E+00	6.40E-01	1.10E-01									
43369	47	3.02E+00	5.40E-01	1.40E-01									
43408	48	3.68E-01	9.20E-02	5.80E-02				.					
43544	49	5.80E+00	8.40E-01	7.60E-02									
99911	50	•			2.10E-01	4.00E-02		5.58E-01	2.80E-01	4.40E-02		•	1.30E+00
			•		•		•		•		1.09E+00	6.30E-01	1.10E+00
43370 44177	51 52	3.71E-01 4.01E+00	1.02E-01 5.40E-01	6.60E-02 6.50E-02		8.30E-01	•	· ·	•	•	-	•	•

USGS				Ra226 (p	Ci/L) - USGS	5			Ra226	6 (pCi/L) - El	PA (quality-	control lab	)
serial	Map key	alp	oha spectro		gan	nma spectro	scopy	alı	oha spectro	scopy	gar	nma spectr	oscopy
number	ĸċy	result	CSU <sup>1</sup>	MDC	result	CSU <sup>1</sup>	na	result	CSU <sup>2</sup>	MDC	result	CSU <sup>2</sup>	MDC
44178	•	3.54E+00	5.20E-01	4.60E-02	· · · ·	· · ·							
99912			•		6.38E+00	5.00E-01					·		
99910 45603	53 54	2.34E+00	3.66E-01	5.80E-02	1.09E+00	2.20E-01		2.98E+00	5.00E-01	2.50E-02	1.36E+00	5.10E-01	6.20E-01
43949		2.34L+00 2.23E+00	3.80E-01	7.10E-02		•	•	2.301+00	J.00L-01	2.30L-02	1.30L+00	J.10L-01	0.202-01
43950	55	2.91E+00	5.00E-01	1.10E-01				· ·					
43833	56	8.75E-01	1.60E-01	6.80E-02									
43546	57	1.12E+00	2.60E-01	8.30E-02									
43552	58	6.33E-01	1.66E-01	9.80E-02									
45428	59	1.76E+00	3.20E-01	8.50E-02	•	•	•	1.53E+00	3.50E-01	2.40E-02	1.63E+00	3.80E-01	4.70E-01
43824	60	6.56E-01	1.22E-01	5.70E-02		•	•	•			·		
43467	61	5.33E-01	1.00E-01	3.50E-02	•	•	•	•	•	•	·	•	•
47127 43472	62 63	3.50E+00 8.96E+00	4.40E-01 1.30E+00	2.50E-02 8.20E-02	•	•	•		•	•	· ·	•	•
99903	64	0.302+00	1.30L+00	0.202-02	2.29E+00	1.80E-01	•	•	·	•	· ·	•	•
44084	65	6.20E-02	6.80E-02	1.10E-01	2.232+00	1.002 01							
44180	66	2.13E-01	5.80E-02	4.10E-02									
44085		3.40E-02	4.20E-02	6.70E-02									
44086	67	4.10E-02	3.20E-02	4.70E-02	6.00E-02	2.00E-01					.		
45087	68	4.39E+00	6.80E-01	4.60E-02	5.40E+00	8.10E-01		4.81E+00	6.30E-01	7.60E-03	4.53E+00	4.40E-01	5.20E-01
45088	•	5.10E+00	6.40E-01	3.10E-02	· · ·	·			· · - ·	·	· ·	· ·	· · ·
45089	69	3.24E+00	4.40E-01	5.10E-02	3.43E+00	5.40E-01		3.05E+00	4.20E-01	1.40E-02	2.72E+00	3.70E-01	4.60E-01
45090		3.78E+00	6.80E-01	1.20E-01			•	•	•	•	· ·	•	•
46849 46850	70	1.97E+00 1.90E+00	3.00E-01 3.20E-02	3.30E-02 3.70E-03	5.60E+00	1.60E-01		•	•	•	·	•	•
40850	71	1.82E-01	3.20E-02 7.60E-02	5.60E-03	•	•	•	•	•	•	· ·	•	•
43622		1.68E-01	8.60E-02	6.80E-02		•	•	•	·	•	· ·	•	•
43623	72	1.57E-01	5.40E-02	4.30E-02			÷						
45224	73	2.88E-01	8.20E-02	5.70E-02				3.59E-01	1.60E-01	2.50E-02			8.30E-01
45223	74	1.79E-01	5.40E-02	4.30E-02				2.51E-01	1.10E-01	1.70E-02	8.43E-01	3.60E-01	7.40E-01
45352	75	1.04E-01	3.60E-02	3.60E-02				6.00E-02	6.40E-02	2.00E-02	.		1.50E+00
											.		1.20E+00
44966	76	1.46E-01	5.60E-02	5.20E-02				5.25E-01	1.90E-01	2.20E-02	3.40E-01	3.30E-01	4.80E-01
					•		•	2.52E-01	1.00E-01	6.80E-03	·	•	•
43832	77	3.06E-01	6.80E-02	3.70E-02	•	•	•	•	·	•	· ·	•	•
44176	78 79	2.34E-01 5.10E-02	6.00E-02 5.60E-02	4.40E-02 8.90E-02	•	•	•		•	•	· ·	•	•
44239 44969	80	8.50E-02	6.40E-02	9.90E-02	7.00E-02	5.00E-02	•	3.41E-01	2.70E-01	6.80E-02	1.43E+00	4.50E-01	5.20E-01
44970		4.40E-01	3.60E-01	5.20E-01	7.001 02	3.00L 02		0.412 01	2.702 01	0.001 02	7.35E-01	5.60E-01	6.90E-01
44971		3.40E-01	2.80E-01	3.90E-01	1.40E-01	4.00E-02	•	1.14E-01	2.60E-01	1.20E-01	1.33E+00	6.20E-01	1.40E+00
44972		2.00E-01	3.80E-01	6.70E-01		•					1.62E+00	4.40E-01	1.30E+00
45220	81	1.63E+00	5.00E-01	3.80E-01	8.20E-01	1.30E-01		1.98E+00	4.40E-01	3.30E-02	6.67E-01	2.90E-01	3.60E-01
45221		1.06E+00	3.00E-01	2.10E-01							.		
99902	82	·	· · ·	· • _ · ·	4.40E-01	1.40E-01			÷	· .	· · ·	•	· · ·
45002	83	4.10E-02	3.40E-02	5.00E-02	•		•	1.96E-01	1.10E-01	9.50E-03	3.25E-01	2.10E-01	6.30E-01
45052		1.49E-01	5.00E-02	5.20E-02	•	•	·	0.005.00	7 405 00	2.005.02			
45051 43948	84 85	1.72E-01 5.40E-01	4.60E-02 2.40E-01	3.50E-02 2.50E-01	•	•		8.60E-02	7.40E-02	2.00E-02	3.35E-01	4.10E-01	7.60E-01
43948	00	3.50E-01	2.40E-01 2.60E-01	2.50E-01 3.40E-01	·	·	·	2.45E-01	1.20E-01	1.70E-02	6.01E-01	3.10E-01	3.80E-01
44175	86	7.50E-01	6.00E-01	6.90E-01	•		•						
99901	87				9.80E-01	8.00E-02							
45348	88	4.55E-01	9.80E-02	5.40E-02				3.77E-01	9.80E-02	7.60E-03	.		7.10E-01
45349		6.51E-01	1.40E-01	5.80E-02							.		
45351	89	1.43E+00	2.20E-01	2.80E-02	1.62E+00	1.30E-01		1.32E+00	3.80E-01	4.10E-02	1.44E+00	3.90E-01	4.80E-01
45350		1.82E+00	2.80E-01	4.30E-02	•					•	.	•	
43411		1.94E+00	4.00E-01	8.10E-02				· ·			·		
44080 44078	90 91	1.63E+00 1.73E+00	3.80E-01	1.30E-01	·	·	·	•	•	•	·	•	•
44078	91 92	1.73E+00 2.89E+00	3.80E-01 4.20E-01	1.30E-01 4.30E-02	•	•	·	•	•	•	· ·	•	•
43428	92	2.89E+00 1.86E+00	4.20E-01 3.80E-01	4.30E-02 1.20E-01	•	•	•		•	•		•	•
43477	93	9.57E+00	1.34E+00	6.40E-02					•	•		•	•
43493		1.01E+01	1.44E+00	5.50E-02									
43969	94	9.06E-02	4.20E-02	5.10E-02							.		
43914	95	4.30E-01	1.34E-01	7.40E-02							.		
43915		4.53E-01	1.28E-01	7.40E-02	•	•	•			•	.		
43410	96	9.99E-02	5.20E-02	6.80E-02							.		
43371	97	8.79E-01	1.70E-01	5.60E-02							•		
43545 43474	98 99	1.61E+00 1.66E+00	4.00E-01 2.40E-01	1.30E-01 4.60E-02	•	·	·	•	•	•	· ·	•	•
40474	33	1.000+00	2.40E-01	4.00E-0Z	•	•	•	•	•	•	•	•	•

USGS	Мар				ci/L) - USGS				L) - EPA (qual	
serial number	key	result	beta countii CSU <sup>3</sup>	ng MDC	gam result	ima spectrosi CSU <sup>1</sup>	copy na	ga result	mma spectr CSU <sup>2</sup>	oscopy MDC
44322	2	5.12E-02	2.90E-01	7.49E-01						
44323					-					
44320										
44321	1	1.75E-01	2.60E-01	6.72E-01						
43372	3	3.37E-01	3.10E-01	7.39E-01						
45344	4	4.35E-01	3.00E-01	5.81E-01	-	-		1.83E-01	2.50E-01	9.70E-01
45345	5	3.92E-01	3.60E-01	8.72E-01					2.002 01	1.30E+00
									•	
45429	6	8.11E-01	4.10E-01	7.93E-01				6.48E-01	7.50E-01	3.40E-01
44087	7	5.32E-01	3.50E-01	7.31E-01	2.90E-01	8.00E-02			•	•
44088		2.52E-01	2.90E-01	7.29E-01			•			
44179	8	5.46E-01	3.30E-01	5.91E-01	-					
44319	9	7.22E-01	3.90E-01	7.68E-01	-					
44328	10	3.12E-01	2.90E-01	6.96E-01	-					
99904	11				9.00E-02	9.00E-02				
45219	12	2.36E-02	2.70E-01	7.68E-01	-					1.00E+00
99909	13		-		1.28E+00	1.60E-01				
99906	14	_		_	9.00E-02	1.50E-01				_
99907	15		-		7.00E-02	7.00E-02			-	
45222	16	2.71E-01	3.30E-01	8.18E-01					•	1.10E+00
43553	17	2.06E-01	2.30E-01	5.23E-01		•	•		•	1.102100
43468	18	8.14E-01	3.60E-01			•	•		•	•
43468 99908	10	0.146-01	J.JUL-UI	5.31E-01	2.30E-01	1.10E-01	•	· ·	•	•
		0 EOE 00	2 00E 01	0 21E 01	2.30E-01	1.10E-01		· ·	•	1 105 00
45066	20	8.58E-02	3.00E-01	8.21E-01	-	-	·	· ·	•	1.10E+00
45426		5.37E-01	3.40E-01	7.29E-01			•			
45425	21	3.92E-01	3.60E-01	8.72E-01	6.70E-01	9.00E-02		9.39E-01	7.20E-01	1.20E+00
45065	22	1.77E-01	3.10E-01	8.15E-01	-	•		1.66E-01	2.60E-01	1.40E+00
45085	23	1.66E-01	2.40E-01	5.63E-01	-		•			1.30E+00
45083		1.56E-01	2.60E-01	6.17E-01			•			
45084	24	2.94E-01	2.80E-01	5.88E-01	-			6.59E-01	7.20E-01	1.10E+00
43473	25	4.69E-01	3.30E-01	7.36E-01	-					
47128	26	2.14E-01	3.50E-01	8.98E-01	7.60E-	01 1.10E-01	Ι.			
47129			-	-	-	-				
44325	27	1.09E+00	4.15E-01	4.97E-01						<u>.</u>
43368	28	1.28E+00	4.30E-01	4.28E-01						_
43475	29	1.97E-01	2.70E-01	6.87E-01	-	-	-		-	-
43407	30	2.45E+00	7.40E-01	7.95E-01	-	•	•		•	•
43831	31	2.43E+00 7.47E-01	4.50E-01	9.33E-01	•	•	•		•	·
99913	51	4.33E-01	4.30E-01 3.70E-01	8.70E-01	-	•	•	•	•	•
	32	4.33E-01 6.60E+00	1.60E+00	8.26E-01	6.40E+00	E 00E 01	•	•	•	-
43951		0.00E+00	1.00E+00	0.20E-UI		5.00E-01			•	•
43968	•	•	-	•	6.61E+00	6.10E-01		•	•	•
44181	•	· · •	•	· · .	· · ·	· ·	•			
44182	33	5.20E+00	1.30E+00	4.91E-01	5.08E+00	5.60E-01			•	•
43845	34		-		-		•			-
43846		3.89E+00	1.10E+00	9.85E-01	4.11E+00	3.80E-01		.		
44329	35	3.64E+00	9.65E-01	7.43E-01	3.84E+00	4.30E-01		.		
44330		3.36E+00	8.90E-01	6.94E-01	-			.	-	
99914	36	2.28E-01	2.90E-01	7.40E-01				.		1.50E+00
44316	37	3.12E-01	3.10E-01	7.16E-01				.		
45125	38	8.33E-01	4.60E-01	8.90E-01				.		9.70E-01
44324	39	4.60E-01	3.50E-01	7.17E-01				.		
44967	40									-
44968		2.52E-01	3.40E-01	8.25E-01	-		•	3.26E-01	2.40E-01	2.40E-01
45424		2.022-01	5. FOL-01	0.202-01	4.19E+01	3.20E+00	•	0.202-01	2.102 01	2.70L-01
45424	41	3.64E+01	7.90E+00	9.04E-01	4.19E+01 3.90E+01	2.94E+00		3.47E+01	2.30E+00	4.50E-01
45423 45422		3.04E+01	7.30E+00	3.04E-01				J.4/E+01	2.300+00	4.JUE-UI
		7 225 01	1 505 01	0 705 01	6.89E+01	5.17E+00		6 705 01	4 105 00	
45421	42	7.23E+01	1.50E+01	8.78E-01	7.89E+01	5.98E+00		6.79E+01	4.10E+00	4.80E-01
99905	43	1.005.01			4.81E+00	5.60E-01		· ·	•	
45343	44	1.99E-01	3.20E-01	8.47E-01	-	-	•	·	•	1.10E+00
45648	45	1.42E-01	3.10E-01	8.37E-01	-	•		.	•	1.50E+00
43409	46	2.15E+00	7.00E-01	8.94E-01			•	.		
43369	47	2.50E+00	7.30E-01	5.12E-01	-	-		.	•	
43408	48	6.04E-01	3.90E-01	8.15E-01	-	-		.		
43544	49	6.03E+00	1.50E+00	5.20E-01				.		
99911	50	5.00E-01	3.60E-01	8.13E-01	5.00E-01	1.10E-01		4.84E-01	7.30E-01	7.30E-01
			•	•	•			.		2.30E+00
	51	1.11E+00	4.70E-01	7.88E-01	-			.	-	
43370					-			· ·	-	•
43370 44177	52	5.77E+00	1.40E+00	7.98E-01	5.57E+00	5.20E-01				

<sup>1</sup>One standard deviation <sup>2</sup>Two standard deviations based on counting error only <sup>3</sup>Two standard deviations

USGS	Мар				Ci/L) - USGS					ity-control lab
serial number	key	result	beta countii CSU <sup>3</sup>	ng MDC	gan result	nma spectro CSU <sup>1</sup>	scopy na	g result	amma spectr CSU <sup>2</sup>	oscopy MDC
99912					6.66E+00	6.40E-01				
99910	53			-	9.70E-01	1.30E-01				
45603	54	6.84E-01	3.70E-01	7.40E-01	•	•		•	•	2.10E+00
43949	·		· · ·	· · · ·		•	•		•	
43950	55	6.35E-01	4.80E-01	1.04E+00						
43833	56	4.40E-01	3.80E-01	8.80E-01						
43546	57	5.05E-01	3.10E-01	6.54E-01						
43552	58	3.70E-01	2.60E-01	5.13E-01	•	•	•		•	
45428	59	3.86E-01	3.40E-01	8.06E-01	•	•	•		•	1,5
43824	60	2.62E-01	2.90E-01	6.37E-01	•	•	•		•	•
43467	61	7.12E-01	3.40E-01	6.08E-01						•
47127	62	3.43E+00	9.50E-01	8.40E-01						
43472	63	1.57E+00	5.20E-01	6.70E-01	4 105 .00		•	•	•	•
99903	64				4.13E+00	2.30E-01		•	•	•
44084	65	2.60E-01	3.10E-01	7.18E-01	•	•	•	•	•	•
44180	66	5.23E-01	3.10E-01	5.51E-01	•	•	•	•	•	•
44085							•	•	•	•
44086	67	3.37E-01	3.20E-01	7.22E-01	1.40E-01	4.00E-02				
45087	68	3.87E+00	1.04E+00	5.94E-01	4.68E+00	4.70E-01		3.40E+00	6.60E-01	2.80E-01
45088		1 775.00	E 40E 01		2 505 . 00	2.005.01	•	2 105.00	E 00E 01	
45089	69	1.77E+00	5.40E-01	4.74E-01	2.59E+00	3.60E-01		2.19E+00	5.80E-01	4.10E-01
45090	70	4.075.00	1.00 - 00		- 	4 205 01	•		•	•
46849	70	4.67E+00	1.20E+00	8.48E-01	5.60E+00	4.30E-01			•	
46850			2.005.01			•	•		•	
43554	71	7.75E-01	3.60E-01	6.47E-01						
43622 43623	70	6.31E-01	0.105.01			•	•		•	
	72		3.10E-01	4.77E-01		•	•		•	1 105 .00
45224	73	1.69E+00	5.70E-01	5.65E-01						1.10E+00
45223	74	8.11E-01	4.40E-01	8.73E-01		•	•		•	1.10E+00
45352	75	4.63E-01	3.60E-01	8.45E-01				1.055.00	1 EOE . 00	2.60E+00
			2.205.01	7 205 01		•	•	1.95E+00	1.50E+00	2.10E+00
44966	76	4.74E-01	3.30E-01	7.38E-01		•	•		•	1.20E+00
43832	77	4.07E-01	4.00E-01	9.15E-01	•	•	•	•	•	-
43632	78	4.07E-01 4.38E-01	4.00E-01 3.00E-01	5.65E-01	•	•	•	•	•	-
44170	78	4.38E-01 4.32E-01	2.70E-01	4.92E-01	•	•	•	•	•	-
44239	80	4.32E-01 9.39E-01	5.30E-01	4.92E-01 1.04E+00	7.60E-01	1.10E-01	•	•	5.80E-01	1.80E+00
44970		3.332-01	J.30L-01	1.04L+00	7.002-01	1.102-01			6.60E-01	2.00E+00
44971	•	1.05E+00	4.90E-01		6.60E-01	9.00E-02	5.00E-01		7.40E-01	2.30E+00
44972		1.032+00	4.502-01	0.702-01	0.002-01	J.00L-02	3.002-01	1.75E+00	1.50E+00	1.80E+00
45220	81	1.70E+00	5.70E-01	7.61E-01	1.48E+00	3.20E-01	5.00E-01	1.02E+00	5.70E-01	2.70E-01
45221		1.702100	3.70L 01	7.012 01	1.402100	0.202 01	5.00L 01	1.022100	3.70E 01	2.702 01
99902	82	-	•	-	4.30E-01	1.10E-01	5.00E-01		•	
45002	83	2.40E-01	2.90E-01	7.28E-01	4.50L-01	1.102-01	5.002-01		•	1.20E+00
45052		2.402-01	2.502-01	7.201-01	•				•	1.202+00
45051	84	1.65E-01	3.10E-01	8.26E-01	•	•	•		•	1.30E+00
43948	85	4.64E-01	3.20E-01	6.29E-01	•	•	•		•	1.002100
45086			0.202 01	0.202 01	•	•	•		•	8.70E-01
44175	86	5.02E-01	3.60E-01	8.05E-01			•		•	
99901	87	5.02E 01	0.002 01	0.032 01	7.50E-01	1.50E-01	•		•	-
45348	88	8.16E-01	3.80E-01	5.88E-01				•	•	1.40E+00
45349		9.43E-01	4.20E-01	7.42E-01						
45351	89	8.35E-01	4.20E-01	7.88E-01	4.00E-01	1.20E-01	•			1.70E+00
45350		0.002 01								
43411		4.34E+00	1.10E+00	7.69E-01	•	•	•		•	-
44080	90	5.02E+00	1.30E+00	5.31E-01	•					•
44078	91	6.60E+00	1.70E+00	6.88E-01	•					•
43428	92				-					
44079		5.90E+00	1.50E+00	7.18E-01	-	•	•			•
43477	93	3.40E-01	2.80E-01	5.71E-01	•		•			
43493		0	2.002 01		•		•			
43969	94	2.34E-01	2.70E-01	6.76E-01	•					•
43914	95	5.96E-01	3.50E-01	7.28E-01	•					
43915					•					
43410	96	3.53E-01	2.80E-01	5.63E-01	•		•			
43371	97	1.18E+00	4.80E-01	7.89E-01	•					•
43545	98	2.41E+00	7.10E-01	5.76E-01	-	•	•		-	•
43474	99	2.99E+00	8.50E-01	6.26E-01	•			l .		•
		2.00L+00	0.000-01	0.202-01	•	•	•	· ·	-	-

<sup>1</sup>One standard deviation <sup>2</sup>Two standard deviations based on counting error only <sup>3</sup>Two standard deviations

USGS		Th22	7 (pCi/L) -	EPA	Th228	(pCi/L) - L	JSGS	Th22	8 (pCi/L) -	EPA	Th23	0 (pCi/L) -	USGS	Th23	0 (pCi/L) -	EPA
serial	Map key	alpha	a spectro	scopy	alpha	spectros	сору	alpha	spectros	сору	alph	a spectro	scopy	alpha	a spectros	сору
number		result	CSU <sup>2</sup>	MDC	result	CSU <sup>1</sup>	MDC	result	CSU <sup>2</sup>	MDC	result	CSU1	MDC	result	CSU <sup>2</sup>	MDC
44320	÷	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
44321 43372	1 3	•	•	•	•	•	•	•	•	•	•	•	•	•	•	
45344	4			÷		÷						÷				
45345	5															
			•	•		•			•	·		·			•	
45429 44087	6 7	-	•	·		•	•		•	•		·	•	•	·	·
44087	/	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
44179	8															
44319	9			•												
44328	10			•		•			•		•				•	
99904 45219	11 12	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
99909	13			÷		÷						÷				
99906	14	-	•			•		•	•							
99907	15			•												
45222	16	•	•			•	•		•	•		·	•	•	•	
43553 43468	17 18	•	•	•	·	•	•	•	•	•	•	•	•	•	•	•
99908	19															
45066	20															
45426		•	•	•	•	•	•	•	•	•		•	•	•	•	
45425 45065	21 22	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
45085	22		•			÷	•		•				•		÷	
45083																
45084	24	-	•	•		•		•	•	•		•	•			
43473	25		•	•		•			•		•				•	
47128 47129	26	•	•	•	•	•		•	•		•	•		•	·	
44325	27															
43368	28	-	•						•							
43475	29			•												
43407	30 31	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
43831 99913	31		•	•		•			•	•	•	•	•		•	•
43951	32				9.70E-01	1.60E-01	4.30E-01				1.80E-01	1.40E-01	4.50E-01			
43968					3.60E-01			-	•			1.60E-01				
44181				•		1.40E-01						1.60E-01				
44182 43845	33 34	•	•	•	7.00E-01 2.10E-01			•	•	•	-3.10E-01	1.50E-01 1.60E-01		•	•	·
43846					2.102 01		4.00L 01				7.002 02					
44329	35	-	•		1.30E-01	1.10E-01	3.80E-01		•		3.00E-01	1.70E-01	5.30E-01			
44330	·				2.80E-01	1.90E-01	6.50E-01				-4.60E-01	1.70E-01	9.40E-01			
99914 44316	36 37	•	•	•	•	•	•	•	•	•	•	•	•	•	•	•
44316	37		•	•		•	•		•	•		•	•		•	•
44324	39		•						•	•						
44967	40		•													
44968	•		•		1.005.00						2005.01					
45424 45423	41	3.88F-01	1.10F-01	6.60E-02	1.83E+00 9.20E-01	2.10E-01 2.80E-01		3.87E+00	2.80E-01	4.60F-02		1.60E-01 2.10E-01		6.41F-02	3.20E-02	2.00F-02
45422	41	0.002-01		J.UUL-UZ	3.36E+00						1.20E-01			0.71L-0Z		2.00L-02
45421	42	7.00E-01	1.40E-01	4.30E-02	3.82E+00			5.18E+00	3.20E-01	4.10E-02	-3.00E-02			6.25E-02	2.80E-02	9.40E-03
99905	43	•	•	•			•	•	•	•			•	•		•
45343 45648	44 45		•	•	· ·		•			•				•		•
43648	45 46		:				•		•				•		:	
43369	47		•						•	•						
43408	48															
43544	49 50		•		2.30E-01	2.30E-01	8.10E-01				3.50E-01	2.00E-01	8.50E-01			
99911	50	•	•	•	· ·	-	•	•	•	•	•	•	•	•	•	
43370	51		•						•							
44177	52															
44178			•						•	•		•		•		
99912	•	•				-										
								-								

USGS	Мар	Th2	27 (pCi/L) ·	- EPA	Th228	8 (pCi/L) - U	SGS	Th22	28 (pCi/L) -	EPA	Th230	(pCi/L) - l	JSGS	Th23	0 (pCi/L) -	EPA
serial number	key	alph result	na spectro CSU <sup>2</sup>	scopy MDC	alpha result	a spectrosc CSU <sup>1</sup>	opy MDC	alph result	a spectros CSU <sup>2</sup>	copy MDC	alpha result	a spectros CSU <sup>1</sup>	copy MDC	alpha result	a spectros CSU <sup>2</sup>	copy MDC
99910	53	····			ioouit			ioouit			ioouit			iooun		
45603	54															
43949								.								
43950 43833	55 56	•	•	•	2.10E-01	2.90E-01	1.00E+00	· ·	•	•	1 505 01	2.10E-01	1 00E 00	· ·	•	·
43633	50 57	•	•	•	2.102-01	2.502-01	1.00E+00		•	•	-1.50E-01	2.10E-01	1.00E+00		·	•
43552	58											-				
45428	59															
43824 43467	60 61	•	·	•		•	•	· ·	•	•		·	·	· ·	·	·
43407 47127	62	•	•	•	5.30E-02	6.10E-02	2.10E-01		·	•	-1.07E-01	7.70E-02	2.70E-01		·	•
43472	63															
99903	64							.								
44084 44180	65 66	•	·	•		•	•	· ·	•	•		·	·	· ·	·	·
44180		•	•	•		•	•		•	•		•	·		·	•
44086	67											-				
45087	68		4.50E-02		1.80E-01	1.60E-01	5.50E-01	1.12E-01	4.40E-02	3.80E-02		1.80E-01		2.90E-02	2.20E-02	2.00E-02
45088 45089	69	9.92E-02 3.97E-03	5.50E-02		1.30E-01	2.30E-01	7.90E-01	1.32E-01	4.60E-02 4.50E-02		1.40E-01	2.30E-01	7.70E-01		1 605 02	9.30E-03
45089 45090	09	3.97E-03 9.97E-03	2.30E-02 2.60E-02		·	•	•		4.50E-02 4.40E-02		·	•	•			9.30E-03 1.60E-02
46849	70				1.49E-01	5.40E-02	1.60E-01	.			6.00E-02	7.20E-02	2.30E-01			
46850					-1.10E-02	6.70E-02	2.40E-01	.			-5.90E-02	6.30E-02	2.20E-01			
43554	71				.			.			.			·		
43622 43623	72	•	•	•		•	•		•	•		•	•		•	•
45224	73															
45223	74							.								
45352	75		·				•		·	•				· ·		·
44966	76		•					· ·	•			•		· ·	•	•
44300		•	:	•		•	•			•		•	•		•	:
43832	77															
44176	78		•	•		•			•	•				.		•
44239 44969	79 80	1.27E+00	/ 10E_01	2 /0F-01	2.10E+01	9.40F-01	8.40E-01	2.06F⊥01	1.40E+00	2 10F-01	1.50E+01	7 20F-01	7 90F-01	1.62F⊥01	1.20E+00	1 10F-01
44970			4.10E 01	2.402 01	2.20E+01		8.80E-01				1.51E+01					
44971		1.34E+00	4.40E-01	2.30E-01	2.10E+01		9.40E-01	2.19E+01	1.50E+00	2.20E-01	1.69E+01			1.71E+01	1.20E+00	1.20E-01
44972		1.005.01		4 005 01	2.07E+01		7.10E-01	1.015.00	2.405.01	2.005.01	1.59E+01			1 005 - 00	2 00F 01	1 205 01
45220 45221	81	1.20E-01	2.50E-01	4.00E-01	1.49E+00 1.61E+00	2.00E-01 1 70E-01	4.90E-01 3.40E-01	1.31E+00	3.40E-01	3.00E-01	1.02E+00	1.90E-01 1.90E-01		1.06E+00	2.80E-01	1.20E-01
99902	82															
45002	83															•
45052 45051	84	•	•	•	·	•	•	· ·	•	•	•	•	·	· ·	•	·
43948	04 85	•	•	•	·	•	•	•	•	•	·	•	•		•	•
45086		-	-	-								-				
44175	86				.			.			.			.		
99901 45348	87 88		•		·		•	·			·			· ·		•
45348 45349	88			•		•			•	•		•	•			
45351	89															
45350	·	•	•	•	.	•	•	·	•	•		•	•	· ·		•
43411 44080	90		•		7.70E-01	1.60E-01	4.20E-01	·			3.60E-01	2 በ∩⊑₋01	6.10E-01	· ·		•
44080	90 91			· ·	4.80E-01	1.30E-01	4.20E-01 3.70E-01			•		1.70E-01				
43428	92				1.15E+00	2.50E-01	7.40E-01	.			-2.00E-01	1.80E-01	9.00E-01			
44079		•	•	•	7.20E-01	1.90E-01	5.70E-01	.	•	•	-1.90E-01	1.50E-01	5.50E-01	.	•	·
43477 43493	93		•		·		•	·			·			· ·		•
43493 43969	94			•		•			•	•		•	•			
43914	95										.					
43915							•	.								
43410	96 07				.			•			.			·		·
43371 43545	97 98	•	•	•	1.00E-02	1.80E-01	6.70E-01		•	•	-8.00F-02	1.80E-01	8.70F-01		•	•
43474	99				1.40E-01	2.10E-01	7.60E-01				0.00E+00					
44322 44323	2				.			.						.		
					·		-	.		•				.		•

USGS	Man	Th23	32 (pCi/L) - l	JSGS	Th2	32 (pCi/L) -	EPA	F	°o210 (pCi/l	L)		Pb210 (pCi/L	.)
serial number	Map key	alpl result	na spectros CSU <sup>1</sup>	copy MDC	alpł result	ia spectros CSU <sup>2</sup>	copy MDC	alpł result	na spectros CSU <sup>3</sup>	scopy MDC	result	beta filtere CSU <sup>3</sup>	d MDC
44322	2							4.52E-03	9.40E-03	2.04E-02	1.13E+00	5.40E-01	9.04E-01
44323						•	•						
44320			•	•		•	•	•	•	•		•	•
44321	1			•		•			1.33E-02	1.79E-02	4.16E-01	3.11E-01	5.90E-01
43372	3		•	•		•	•	-4.71E-03		4.95E-02	1.98E-01	3.70E-01	8.03E-01
45344	4		•	•		•	•	2.46E-03	1.30E-02	3.88E-02	4.26E-01	3.60E-01	6.90E-01
45345	5		•	•		•	•	4.86E-03	1.20E-02	2.91E-02	5.59E-01	4.60E-01	9.09E-01
45429	6			•		•	•	3.84E-01	1.10E-01	3.13E-02	4.14E-01	3.60E-01	7.03E-01
44087	7		•	•	•	•		2.23E-02	1.80E-02	1.91E-02	4.46E-01	3.00E-01 3.00E-01	5.70E-01
44088								1.77E-02	1.80E-02	1.20E-02	2.91E-01	2.90E-01	5.84E-01
44179	8							6.27E-03	1.10E-02	2.21E-02	5.86E-01	3.00E-01	5.31E-01
44319	9							1.03E-02	1.90E-02	3.64E-02	-3.75E-01	4.50E-01	1.03E+00
44328	10							1.84E-02	2.10E-02	1.66E-02	4.28E-01	3.30E-01	6.30E-01
99904	11		•	•		•	•	6.26E-02	3.60E-02	2.34E-02	4.75E-01	4.50E-01	9.14E-01
45219	12		•	•			•	2.34E-02	1.90E-02	1.06E-02	3.95E-01	3.60E-01	7.09E-01
99909	13		•	•		•	•	1.24E+00	2.49E-01	1.79E-02	4.14E+00	9.60E-01	6.13E-01
99906	14		•	•		•	•	5.75E-01 9.55E-01	1.22E-01 1.66E-01	2.06E-02	1.75E+00 2.60E+00	5.10E-01 6.78E-01	5.94E-01
99907 45222	15 16	· ·	•	•	•	•	•			1.04E-02 2.21E-02			6.41E-01
43222	16 17	· ·	•	•	•	•	•	1.27E-01 1.75E-02	4.50E-02 1.60E-02	2.21E-02 1.77E-02	2.13E+00 2.71E-01	6.10E-01 3.00E-01	6.73E-01 6.11E-01
43553	17	· ·	•	•		•		1.01E-02	1.50E-02 1.50E-02	2.65E-02	2.71E-01 2.72E-02	4.30E-01	0.11E-01 1.01E+00
99908	19							1.33E-02	1.80E-02	2.88E-02	5.98E-01	4.40E-01	8.45E-01
45066	20							3.92E-03	7.90E-03	1.06E-02	1.35E+00	4.70E-01	6.50E-01
45426								-1.37E-03		3.28E-02	-7.70E-01	1.10E+00	2.53E+00
45425	21							4.86E-03	1.20E-02	2.91E-02	7.95E-01	7.30E-01	1.48E+00
45065	22							1.42E-03	7.50E-03	2.25E-02	1.49E-01	3.20E-01	6.69E-01
45085	23			•		•		0.00E+00	1.10E-02	9.90E-03	5.85E-01	3.40E-01	6.11E-01
45083			•	•		•	•	-4.04E-03		4.25E-02	1.94E+00	8.40E-02	1.34E+00
45084	24		•	•		•	•	-7.92E-04		1.90E-02	8.23E-01	4.00E-01	6.76E-01
43473 47128	25 26		·	•	•	•	·	2.78E-02	2.60E-02 1.50E-02	2.77E-02 8.96E-03	1.50E-01 3.00E-01	3.50E-01 3.20E-01	7.86E-01 6.71E-01
47128	20		•	•	•	•	•	1.00E-02	1.30E-02	0.30E-03	3.00E-01	3.20E-01	0.712-01
44325	27		•	•		•	•	2.17E-02	2.73E-02	3.71E-02	6.08E-01	3.70E-01	6.77E-01
43368	28							1.49E-02	1.60E-02	1.87E-02	3.74E-01	4.00E-01	8.31E-01
43475	29							7.36E-02	4.00E-02	1.42E-02	-5.95E-02	3.10E-01	7.34E-01
43407	30							1.08E-02	1.30E-02	1.84E-02	3.99E-01	4.40E-01	9.11E-01
43831	31							1.60E-02	1.40E-02	8.63E-03	5.77E-01	3.10E-01	5.45E-01
99913			•	•		•		2.73E-02	2.10E-02	2.03E-02	6.02E-01	3.29E-01	5.90E-01
43951	32	2.50E-02	2.20E-02	7.50E-02		•	•	4.69E-03	9.40E-03	1.27E-02	2.75E-01	2.60E-01	5.30E-01
43968		5.10E-02	3.00E-02	4.60E-02		•	·		•	•	· ·	•	•
44181		2.00E-02	2.80E-02	1.10E-01		•	•		1.20E-02	2 22F 02	5.31E-01	2.205.01	
44182 43845	33 34	5.50E-02 -1.10E-02	4.10E-02 4.80E-02	1.40E-01 2.10E-01	•	•	•	6.59E-03	1.20E-02	2.33E-02	5.31E-01	3.20E-01	5.96E-01
43846	54	-1.102-02	4.00L-02	2.102-01	•	•	•	6.90E-03	1.80E-02	4.13E-02	1.67E-01	2.70E-01	5.80E-01
44329	35	8.00E-03	3.00E-02	1.30E-01		•			1.96E-02	4.13E-02 1.76E-02	4.03E-01	3.38E-01	6.56E-01
44330		4.00E-02	2.80E-02	5.40E-02					2.00E-02	1.35E-02	1.90E-01	4.40E-01	9.72E-01
99914	36								3.00E-03	3.61E-02	6.01E-01	5.40E-01	1.03E+00
44316	37								1.30E-02	7.58E-03	-2.85E-01	6.00E-01	1.41E+00
45125	38	.	•	•		•			1.10E-02	2.53E-02	6.55E-01	3.70E-01	6.60E-01
44324	39	·	•	•	•	•		9.21E-03	1.31E-02	1.25E-02	6.27E-01	3.62E-01	6.49E-01
44967	40	·	•	•		•	·		0.007.00				
44968	•		1 005 00	2 405 00	•	•	•	0.00E+00	3.20E-02	2.69E-02	4.45E-01	4.10E-01	8.16E-01
45424		2.50E-02	1.80E-02	3.40E-02			1 20E 02	1.005.00		1 605 00	2.765.01	4 00E 01	0.000.01
45423	41	0.00E+00	2 005 02	4.90E-02	1.92E-02	1.70E-02	1.20E-02	1.23E-02	1.70E-02	1.66E-02	2.76E-01	4.00E-01	8.63E-01
45422 45421	42	-4.70E-02		1.90E-01	6.24E-03	8.80E-03	9.40E-03	3 205 02	2.50E-02	2.08E-02	4.88E-01	3.70E-01	7.00E-01
45421 99905	42 43	-1.60E-02	2.00E-02	1.40E-01	0.24E-U3	0.0UE-U3	J.40E-UJ		2.50E-02 1.50E-02	2.08E-02 1.20E-02	4.88E-01 2.65E-01	3.70E-01 4.10E-01	7.00E-01 8.77E-01
45343	43 44	· ·		•		•			1.50E-02 2.00E-02	1.20E-02 1.79E-02	2.05E-01 2.98E-01	4.10E-01 3.20E-01	6.48E-01
45648	44		•			•			1.90E-02	1.77E-02	9.44E-01	4.10E-01	6.65E-01
43409	46								8.00E-02	1.08E-02	7.00E-01	4.90E-01	9.34E-01
43369	47								1.50E-02	1.16E-02	4.38E-01	4.50E-01	9.30E-01
43408	48								2.20E-02	2.96E-02	1.95E-01	3.50E-01	7.75E-01
43544	49	6.60E-02	3.30E-02	4.40E-02		•			2.60E-02	9.18E-03	4.96E-01	3.50E-01	6.71E-01
99911	50	.							1.80E-02	1.65E-02	-6.90E-04	5.37E-01	8.30E-01
		.	•	•		•			•	•		•	•
43370	51	.						· ·	· · · ·		5.09E-01	4.20E-01	8.30E-01
44177	52	.	•	•		•		9.30E-02	3.80E-02	1.73E-02	6.30E-01	3.00E-01	5.02E-01
44178							-						

<sup>1</sup>One standard deviation <sup>2</sup>Two standard deviations based on counting error only <sup>3</sup>Two standard deviations

USGS		Th2	:32 (pCi/L) -	USGS	Th	232 (pCi/L) -	EPA		Po210 (pCi/	L)		Pb210 (pCi/	L)
serial number	Map key	alp result	ha spectros CSU <sup>1</sup>	scopy MDC	alp result	ha spectros CSU <sup>2</sup>	scopy MDC	alp result	ha spectro: CSU <sup>3</sup>	scopy MDC	result	beta filtere CSU <sup>3</sup>	d MDC
99912		rooure			rooun			1.10E-01	4.50E-02	2.03E-02	1.07E+00	4.00E-01	5.90E-01
99910	53	•	•	•	· ·	•	•	2.16E-01	4.50E-02 7.00E-02	2.03E-02 2.73E-02	1.10E+00	4.00E-01 5.30E-01	8.81E-01
45603	50 54							8.27E-03	1.50E-02	2.92E-02	1.102100	5.00L 01	0.012 01
43949													
43950	55							1.24E-02	1.80E-02	1.68E-02	2.76E-01	2.60E-01	5.15E-01
43833	56	2.50E-02	2.50E-02	6.80E-02				1.89E-02	2.10E-02	2.97E-02	7.59E-01	3.30E-01	5.25E-01
43546	57							1.49E-02	1.90E-02	2.54E-02	4.42E-01	3.20E-01	6.12E-01
43552	58	•	•	•	•	•	•	6.22E-03	8.80E-03	8.42E-03	5.74E-01	3.10E-01	5.41E-01
45428	59 60	•	•	·	•	•	•	0.00E+00	2.00E-02	1.77E-02	4.18E-01	3.50E-01	6.83E-01
43824 43467	60 61	•	•	·	· ·	•	•	0.00E+00 0.00E+00	1.00E-02 1.40E-02	9.34E-03 3.47E-02	4.28E-01 2.10E-01	2.90E-01 3.40E-01	5.49E-01
43407	62	1.10E-02	1.10E-02	2.90E-02	· ·	•	•	3.05E-02	2.20E-02	3.47E-02 1.87E-02	7.57E-01	3.40E-01 3.80E-01	7.37E-01 6.28E-01
43472	63			2.002 02				8.25E-03	1.50E-02	2.91E-02	5.44E-01	4.00E-01	7.80E-01
99903	64							2.92E-02	1.90E-02	1.42E-02	7.99E-01	5.30E-01	1.01E+00
44084	65							9.95E-03	1.40E-02	1.35E-02	9.20E-01	3.70E-01	5.50E-01
44180	66							1.06E-02	1.30E-02	1.82E-02	8.68E-01	3.40E-01	5.19E-01
44085							•	•		•	•	•	•
44086	67							4.40E-03	8.80E-03	1.19E-02	5.30E-01	3.10E-01	5.62E-01
45087	68	8.80E-02	3.60E-02	4.00E-02	0.00E+00	1.10E-02	1.10E-02	2.70E-03	9.35E-03	2.54E-02	1.29E+00	4.58E-01	6.52E-01
45088 45089	69	-5.70E-02	4.90E-02	2.80E-01	6.18E-03	1.10E-02	2.10E-02	0.00E+00	1.80E-02	1.64E-02	4.97E-01	3.60E-01	6.87E-01
45089			•	•	2.07E-03	6.50E-02	1.60E-02	0.00E+00	1.00E-02	1.04E-02	4.572-01	3.00E-01	0.0/E-01
46849	70	7.00E-04	2.30E-03	1.90E-02		0.00L-00		9.70E-03	1.10E-02	8.70E-03	1.21E-01	2.84E-01	6.32E-01
46850		1.90E-03	8.20E-03	4.20E-02									
43554	71							2.23E-03	7.70E-03	2.10E-02	6.47E-01	3.20E-01	5.48E-01
43622	•		•	•		•	•	•	•	•		•	•
43623	72	•	•		•	•	•	9.62E-03	1.30E-02	2.09E-02	2.91E-01	3.40E-01	7.18E-01
45224	73		•	•		•	•	7.75E-03	1.60E-02	3.49E-02	1.16E+00	4.30E-01	6.25E-01
45223	74	•	•	·	•	•	•	1.68E-02	2.00E-02	1.52E-02	7.97E-01	3.60E-01	5.97E-01
45352	75	•	•	•	•	•	•	8.06E-03	1.50E-02	2.84E-02	2.10E-02	3.90E-01	9.05E-01
44966	76	·	•	·	·	•	•	-9.32E-04	1.90E-03	2.23E-02	4.90E-01	3.50E-01	6.70E-01
43832	77							5.11E-02	3.40E-02	2.49E-02	1.03E+00	3.80E-01	5.61E-01
44176	78							1.90E-02	1.90E-02	1.28E-02	4.79E-01	3.00E-01	5.65E-01
44239	79					•	•	6.53E-02	3.70E-02	1.36E-02	7.29E-01	3.50E-01	5.94E-01
44969	80				1.62E+01	1.20E+00	4.70E-02	•	•	•		•	•
44970 44971	•	1.58E+01 1.77E+01	8.20E-01	1.80E-01 5.50E-02	1.51E+01	1 205 00	5.10E-02	•	•	•		•	•
44971		1.65E+01	8.80E-01 7.40E-01	1.20E-02	1.512+01	1.20E+00	J.10E-02	•	•	•	•	•	•
45220	81	8.70E-01	1.20E-01	1.20E-01	1.08E+00	2.80E-01	1.10E-01	4.79E-02	3.40E-02	1.62E-02	5.43E-01	3.90E-01	7.45E-01
45221		1.16E+00	1.20E-01	1.10E-01									
99902	82							2.22E-02	1.80E-02	1.56E-02	5.00E-01	5.13E-01	9.70E-01
45002	83							1.66E-02	1.60E-02	2.04E-02	5.60E-01	3.70E-01	6.94E-01
45052					.				•		· ·	· - · -	·
45051	84	· ·	•	•		•	•	4.99E-02	2.70E-02	9.64E-03	5.53E-01	3.70E-01	7.02E-01
43948	85	•	•	•	•	•	•	3.07E-03	7.80E-03	1.84E-02	4.69E-01	3.00E-01	5.66E-01
45086 44175	86	· ·	•	•	· ·	•	•	3.13E-03	6.30E-03	8.48E-03	2.74E-01	2.90E-01	6.01E-01
99901	80 87	·	•	•	i i	•	•	2.37E-03	0.30E-03 1.90E-02	2.03E-03	8.13E-01	5.83E-01	5.80E-01
45348	88							8.44E-03	1.20E-02	1.14E-02	7.84E-01	4.70E-01	8.61E-01
45349	•							3.60E-03	1.20E-02	3.39E-02	6.43E-01	3.70E-01	6.66E-01
45351	89				.			1.59E-02	1.60E-02	1.08E-02	5.72E-01	4.00E-01	7.42E-01
45350		.			.						· ·		
43411						•	•				1.80E-01	4.00E-01	8.83E-01
44080	90	7.00E-03	3.00E-02	1.40E-01	.	•	•	6.38E-02	3.60E-02	2.21E-02	8.15E-01	3.40E-01	5.48E-01
44078	91 02	1.30E-02	3.80E-02	1.50E-01	· ·		•	5.03E-03	1.00E-02	1.36E-02	4.19E-01	3.00E-01	5.79E-01
43428 44079	92	9.50E-02 1.50E-02	4.30E-02 1.50E-02	5.10E-02 4.10E-02	· ·	•	•	-1.84E-03	2.60E-03	2.60E-02	5.41E-01	2.90E-01	5.19E-01
43477	93	1.002-02	1.502-02	4.10E-02		•	•	9.57E-02	3.90E-03	2.00E-02 1.71E-02	7.14E-01	4.20E-01	7.73E-01
43493											.		
43969	94				.			4.85E+00	6.10E-01	1.53E-02	2.39E-01	2.90E-01	6.10E-01
43914	95							5.61E-03	8.00E-03	7.59E-03	4.33E-01	3.10E-01	6.03E-01
43915	•		•			•				•	.		•
43410	96	· ·	•		· ·	•		1.96E-03	6.80E-03	1.85E-02	1.79E-01	3.80E-01	8.46E-01
43371	97		Э.СОГ ОЭ		.	•	•	0.705.00		1.005.00	8.79E-01	4.50E-01	7.65E-01
43545	98 99	3.70E-02	2.60E-02	5.00E-02	·	•		2.78E-03	7.10E-03	1.66E-02	6.34E-01	3.00E-01 3.70E-01	5.04E-01
43474	ษษ	1.90E-02	1.90E-02	5.20E-02	·	•	•	1.33E-02	1.40E-02	1.68E-02	3.34E-01	3.70E-01	7.76E-01
<sup>1</sup> One sta													

<sup>1</sup>One standard deviation <sup>2</sup>Two standard deviations based on counting error only <sup>3</sup>Two standard deviations