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Occurrence and Hydrogeochemistry of Radiochemical Constituents in Groundwater of Jefferson County and Surrounding Areas, Southwestern Montana, 2007 through 2010

Scientific Investigations Report 2013–5235

U.S. Department of the Interior
U.S. Geological Survey

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By Rodney R. Caldwell, David A. Nimick, and Rainie M. DeVaney

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SALLY JEWELL, Secretary

U.S. Geological Survey
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Conversion Factors and Datums

Multiply	By	To obtain
Length		
inch (in.)	25,400	micrometer (μm)
inch (in.)	25.4	millimeter (mm)
inch (in.)	2.54	centimeter (cm)
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)
Area		
acre	4,047	square meter (m^2)
square mile (mi^2)	2.590	square kilometer (km^2)
Volume		
gallon (gal)	3.785	liter (L)
Radioactivity		
picocurie per liter (pCi/L)	0.037	becquerel per liter (Bq/L)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$°F= (1.8×°C) +32$$

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows:

$$°C= (°F-32)/1.8$$

Vertical coordinate information is referenced to the North American Vertical Datum of 1988 (NAVD 88).

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Altitude, as used in this report, refers to distance above specified vertical datum.

Water-Quality Units

[Used in this report]

Ci	curie
mg/L	milligrams per liter
pCi/L	picocuries per liter
μg/L	micrograms per liter
μm	micrometer
μS/cm	microsiemens per centimeter at 25 degrees Celsius

Abbreviations

AMCL	U.S. Environmental Protection Agency alternative maximum contaminant level
ASTM	American Society for Testing and Materials
Bq	becquerel
$CaCO_3$	calcium carbonate
CA-MCL	California maximum contaminant level
C_{env}	concentration of environmental sample
C_{rep}	concentration of replicate sample
CSU	combined standard uncertainty
E	east
ET	evapotranspiration
Fe^{2+}	iron
ft	feet
GPS	global positioning system
GWIC	Montana Bureau of Mines and Geology Groundwater Information Center
HASL	Health and Safety Laboratory
HNO_3	nitric acid
LHA	U.S. Environmental Protection Agency lifetime health advisory
LRL	laboratory reporting level
LT-MDL	long–term method detection limit
MBMG	Montana Bureau of Mines and Geology
MCL	U.S. Environmental Protection Agency maximum contaminant level
MCLG	U.S. Environmental Protection Agency maximum contaminant level goal
MDC	minimum detectable concentration
mi^2	square miles
MMM	multimedia mitigation
Mn^{2+}	manganese
mrem/yr	millirems per year
MRL	minimum reporting level
N	north
NA	not applicable
NAS-NS	National Academy of Sciences-Nuclear Science Series
NAWQA	U.S. Geological Survey National Water Quality Assessment program
NELAC	National Environmental Laboratory Accreditation Conference
ND	not detected

NO_3^-	nitrate
NWIS	U.S. Geological Survey National Water Information System
NWQL	U.S. Geological Survey National Water Quality Laboratory
p	attained statistical significance level
redox	reduction/oxidation
RPD	relative percent difference
SDWR	U.S. Environmental Protection Agency secondary drinking water regulation
SO_4^{2-}	sulfate
ssLc	sample-specific reporting level
ssMDC	sample-specific minimum detectable concentration
USEPA	U.S. Environmental Protection Agency
USGS	U.S. Geological Survey
yr	years

Symbols

α	Acceptable statistical significance level for rejecting null hypothesis
λ	Decay constant, half-life
KЄu	Cretaceous through Cambrian undifferentiated rocks
p	Attained statistical significance level
pЄu	Precambrian undifferentiated rocks
Qs	Quaternary sediment
TKi	Tertiary through Cretaceous igneous intrusive rocks
TKv	Tertiary through Cretaceous volcanic rocks
Ts	Tertiary sediment

Acknowledgments

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Abstract

The U.S. Geological Survey, in cooperation with Jefferson County and the Jefferson Valley Conservation District, sampled groundwater in southwestern Montana to evaluate the occurrence and concentration of naturally-occurring radioactive constituents and to identify geologic settings and environmental conditions in which elevated concentrations occur. A total of 168 samples were collected from 128 wells within Broadwater, Deer Lodge, Jefferson, Lewis and Clark, Madison, Powell, and Silver Bow Counties from 2007 through 2010. Most wells were used for domestic purposes and were primary sources of drinking water for individual households. Water-quality samples were collected from wells completed within six generalized geologic units, and analyzed for constituents including uranium, radon, gross alpha-particle activity, and gross beta-particle activity. Thirty-eight wells with elevated concentrations or activities were sampled a second time to examine variability in water quality throughout time. These water-quality samples were analyzed for an expanded list of radioactive constituents including the following: three isotopes of uranium (uranium-234, uranium-235, and uranium-238), three isotopes of radium (radium-224, radium-226, and radium-228), and polonium-210. Existing U.S. Geological Survey and Montana Bureau of Mines and Geology uranium and radon water-quality data collected as part of other investigations through 2011 from wells within the study area were compiled as part of this investigation. Water-quality data from this study were compared to data collected nationwide by the U.S. Geological Survey through 2011.

Radionuclide samples for this study typically were analyzed within a few days after collection, and therefore data for this study may closely represent the concentrations and activities of water being consumed locally from domestic wells. Radioactive constituents were detected in water from every well sampled during this study regardless of location or geologic unit. Nearly 41 percent of sampled wells had at least one radioactive constituent concentration that exceeded U.S. Environmental Protection Agency drinking-water standards or screening levels. Uranium concentrations were higher than the U.S. Environmental Protection Agency maximum contaminant level (MCL) of 30 micrograms per liter in samples from 14 percent of the wells. Radon concentrations exceeded a proposed MCL of 4,000 picocuries per liter in 27 percent of the wells. Combined radium (radium-226 and radium-228) exceeded the MCL of 5 picocuries per liter in samples from 10 of 47 wells. About 40 percent (42 of 104 wells) of the wells had gross alpha-particle activities (72-hour count) at or greater than a screening level of 15 pCi/L. Gross beta-particle activity exceeded the U.S. Environmental Protection Agency 50 picocuries per liter screening level in samples from 5 of 104 wells. Maximum radium-224 and polonium-210 activities in study wells were 16.1 and 3.08 picocuries per liter, respectively; these isotopes are constituents of human-health concern, but the U.S. Environmental Protection Agency has not established MCLs for them.

Radioactive constituent concentrations or activities exceeded at least one established drinking-water standard, proposed drinking-water standard, or screening level in groundwater samples from five of six generalized geologic units assessed during this study. Radioactive constituent concentrations or activities were variable not only within each geologic unit, but also among wells that were completed in the same geologic unit and in close proximity to one another. Established or proposed drinking-water standards were exceeded most frequently in water from wells completed in the generalized geologic unit that includes rocks of the Boulder batholith and other Tertiary through Cretaceous igneous intrusive rocks (commonly described as granite). Specifically, of the wells completed in the Boulder batholith and related rocks sampled as part of this study, 24 percent exceeded the MCL of 30 micrograms per liter for uranium, 50 percent exceeded the proposed alternative MCL of 4,000 picocuries per liter for radon, and 27 percent exceeded the MCL of 5 micrograms per liter for combined radium-226 and radium-228.

Elevated radioactive constituent values were detected in samples representing a large range of field properties and water types. Correlations between radioactive constituents and

pH, dissolved oxygen, and most major ions were not statistically significant (*p*-value > 0.05) or were weakly correlated with Spearman correlation coefficients (rho) ranging from -0.5 to 0.5. Moderate correlations did exist between gross beta-particle activity and potassium (rho = 0.72 to 0.82), likely because one potassium isotope (potassium-40) is a beta-particle emitter. Total dissolved solids and specific conductance also were moderately correlated (rho = 0.62 to 0.71) with gross alpha-particle and gross beta-particle activity, indicating that higher radioactivity values can be associated with higher total dissolved solids.

Correlations were evaluated among radioactive constituents. Moderate to strong correlations occurred between gross alpha-particle and beta-particle activities (rho = 0.77 to 0.96) and radium isotopes (rho = 0.78 to 0.92). Correlations between gross alpha-particle activity (72-hour count) and all analyzed radioactive constituents were statistically significant (*p*-value < 0.05), and therefore, gross alpha-particle activity likely may be used as a screening tool for determining the presence of radionuclides in area waters. In this study, gross alpha-particle activities of 7 picocuries per liter or greater were associated with all radioactive constituents whose concentrations exceeded drinking-water standards or screening levels.

Radiochemical results varied temporally in samples from several of the thirty-eight wells sampled at least twice during the study. The time between successive sampling events ranged from about 1 to 10 months for 29 wells to about 3 years for the other 9 wells. Radiochemical constituents that varied by greater than 30 percent between sampling events included uranium (29 percent of the resampled wells), and radon (11 percent of the resampled wells), gross alpha-particle activity (38 percent of the resampled wells), and gross beta-particle activity (15 percent of the resampled wells). Variability in uranium concentrations from two wells was sufficiently large that concentrations were less than the MCL in the first set of samples and greater than the MCL in the second.

Sample holding times affect analytical results in this study. Gross alpha-particle and gross beta-particle activities were measured twice, 72 hours and 30 days after sample collection. Gross alpha-particle activity decreased an average of 37 percent between measurements, indicating the presence of short-lived alpha-emitting radionuclides in these samples. Gross beta-particle activity increased an average of 31 percent between measurements, indicating ingrowth of longer-lived beta-emitting radionuclides.

Introduction

Groundwater is the primary source of drinking water for most rural residents and many municipalities of Jefferson County and surrounding areas in southwestern Montana (fig. 1). Residents and government officials are concerned about the quantity and quality of water supplies in the area. One specific concern relates to the presence of radioactive

constituents in groundwater. Elevated concentrations or activities of radon, radium, gross alpha-particles, and gross beta-particles have been documented previously in area groundwater (Leonard and Janzer, 1978; Clark and Briar, 1993; Miller and Coffey, 1998). Recent, but sparse, data available through monitoring of public water-supply systems and private wells indicated that uranium and other naturally-occurring radioactive constituents are present, in some places, at levels of human-health concern. One reason for the concern about radioactive constituents in groundwater is the presence of the Boulder batholith, which underlies parts of Jefferson County and surrounding areas. Uranium and thorium concentrations are elevated in granitic rocks of the Boulder batholith (Tilling and Gottfried, 1969). Uranium and thorium, as well the radioactive constituents produced from the radioactive decay of those constituents, could be transported in the groundwater system.

The rapid increase in population and residential development in Jefferson County (U.S. Census Bureau, 2013) and surrounding areas translates to greater overall reliance on groundwater and to increased use of groundwater from previously less developed areas, such as foothills and mountainous areas, where groundwater is withdrawn from bedrock. However, little information is available to assess the presence or absence of radioactive constituents. In response to this information gap, the U.S. Geological Survey (USGS), in cooperation with Jefferson County and the Jefferson Valley Conservation District, initiated a multiyear investigation to assess the quality of groundwater within the county and surrounding areas, with an emphasis on documenting the occurrence and source of radioactive constituents of known human-health concern. During the study, water-quality data from existing wells in areas underlain by the Boulder batholith and other geologic units were collected and compiled. These data were then examined to determine the distribution of radioactive constituents in groundwater in relation to location, hydrogeologic unit, and geochemical conditions conducive to mobilization of these elements.

Purpose and Scope

This report describes the occurrence of radiochemical constituents, namely uranium, radon, gross alpha- and beta-particle activity, radium, and polonium, in groundwater samples in Jefferson County and surrounding areas of Montana. Samples were collected from 128 water wells from 2007 through 2010. Historical data collected by other agencies also are discussed. The methods used for sample collection and analysis are described, and quality control results are summarized. Concentrations of radiochemical and ancillary constituents are described, detailed in tables, and are compared to drinking-water standards established by the USEPA. The geographic distribution and geologic units of the sampled wells are described. Geological, hydrological, and geochemical factors that could affect the occurrence of radiochemical constituents are evaluated and discussed.
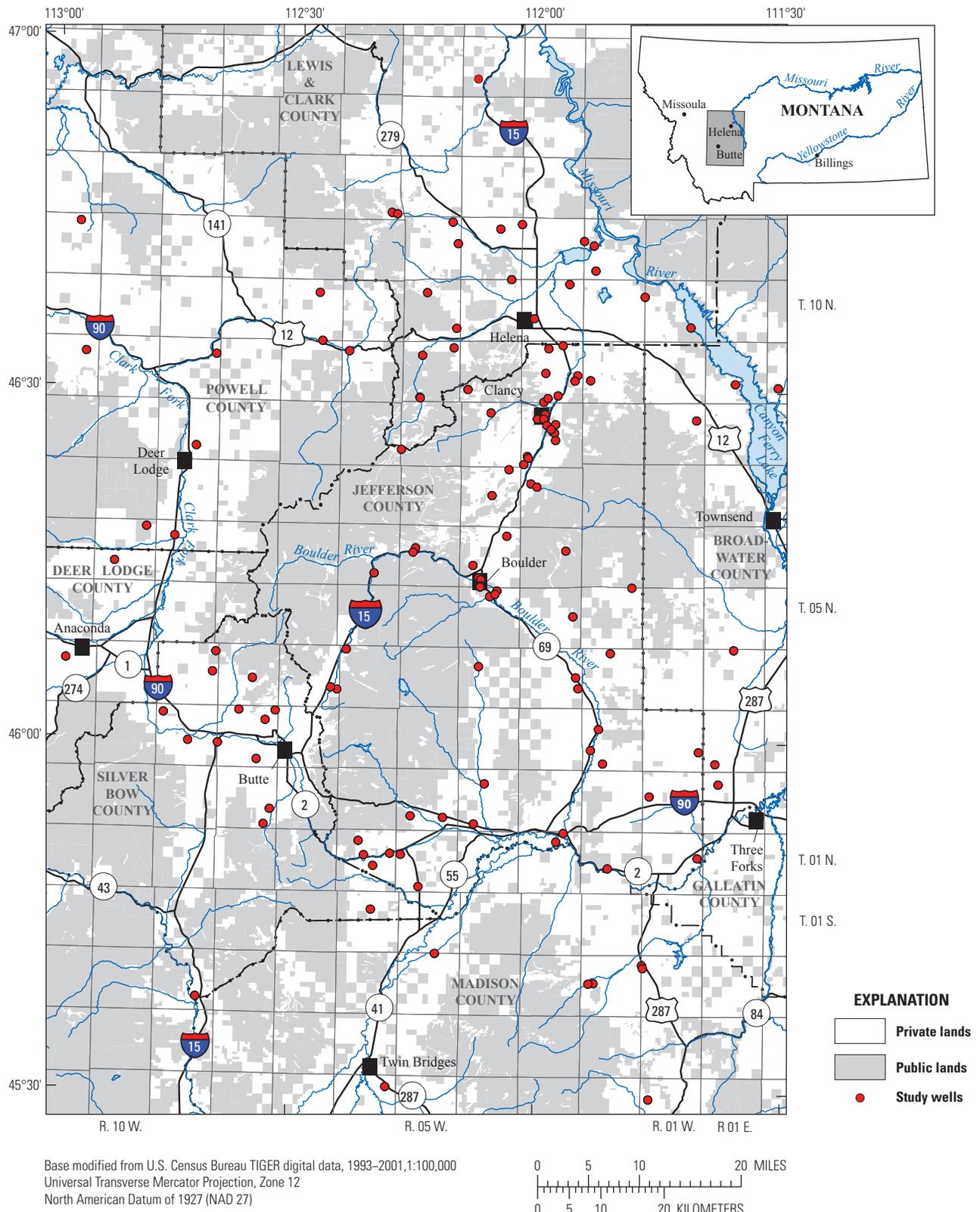
Figure 1.   Location of study area and sampled wells in Jefferson County and surrounding areas, Montana.

General Radiochemistry

Radioactivity describes a group of processes by which matter and energy are released from the nucleus of unstable (radioactive) isotopes or radionuclides, as alpha particles, beta particles, or gamma rays (McCurdy and others, 2008). The radiation that is emitted can strip electrons from atoms with which it collides, causing atoms to become ionized (Szabo and others, 2005). Naturally radioactive constituents are found as trace elements in most rocks and soils and are formed principally by the radioactive decay of uranium-238 and thorium-232 (Zapecza and Szabo, 1988). The radioactive decay of uranium and thorium gives rise to numerous decay products (progeny) or radiogenic daughter products. For example, during decay of uranium, alpha and beta particles are emitted and radium and radon are formed as interim radiogenic daughter products; these, in turn, ultimately decay to a stable form of lead (fig. 2).

Radioactive constituents present in water are colorless, odorless, and tasteless and typically cannot be detected by our senses. The occurrence and distribution of radioactive constituents in groundwater are dependent upon the local geology, specifically the presence and solubility of elements (Zapecza and Szabo, 1988). Chemical processes such as mineral dissolution, desorption, ion exchange, and alpha recoil control the release of radioactive constituents as groundwater travels through pore spaces and fractures within aquifers and interacts with the aquifer matrix (solid material). In order for radiogenic daughter products to be present, the parent radionuclide must be present in the local aquifer. Movement of each radionuclide is dependent on its solubility in water (Zapecza and Szabo, 1988). Alpha recoil is a process in which kinetic energy is imparted to the product nuclide during alpha-particle decay, damaging the crystal lattice surrounding the atom and allowing the atom to break free from the solid structure (Szabo and others, 2005). Through recoil in the opposite direction from the emitted alpha particle, radiogenic daughter products may be directly "ejected" into the pores or eventually may be dissolved at the locus of the crystal-lattice damage depending on the proximity of the parent radionuclide to the mineral-grain boundary (Szabo and others, 2005).

Parent and daughter radionuclides can occur in a state of secular equilibrium in which the activities of all radionuclides within a single decay series are nearly equal. Two conditions are necessary for secular, or long lasting, equilibrium: (1) activity of the progeny eventually equals that of the parent and the parent radionuclide must have a half-life (λ) much longer (at least 50 times) than that of any other radionuclide in the series and (2) a sufficiently long period of time must have elapsed (for example 10 half-lives of the decay product having the longest half-life) to allow for ingrowth of the decay products (Argonne Laboratories, 2007). Combined activity of parent and daughter radionuclides after secular equilibrium is exactly twice that of the original parent; however, concentrations of radiogenic daughter products are typically much higher in natural waters than expected with secular equilibrium.

The main alpha-emitting substances in water are decay products of uranium and thorium. Uranium is the heaviest element that commonly occurs in low concentrations in soil, rock, surface water, and groundwater (Argonne National Laboratory, 2007). Most radionuclides belong to the uranium-238 radioactive decay series (fig. 2). Uranium-238, the most abundant natural isotope of uranium, is a long-lived radionuclide with a half-life of 4.49 billion years, decaying very slowly by emitting alpha particles (fig. 2). In a typical sample of natural uranium, almost all of the mass consists of the uranium-238 isotope (99.27 percent). Less than 1 percent of the mass consists of uranium-235 and uranium-234. Radiogenic daughter products, those produced by radioactive decay, include uranium-234, radium-226, radon-222, and polonium-210, all of which were analyzed in groundwater sampled during this study. Uranium can enter the hydrologic cycle through the weathering or dissolution of uranium-bearing minerals. Uranium is soluble in oxidized waters, but is much less soluble in reduced conditions owing to precipitation of uraninite (Langmuir, 1978). Uranium also tends to be strongly adsorbed onto humic substances (Focazio and others, 2001).

Radium is derived from uranium and thorium and has three important isotopes: radium-224, radium-226, and radium-228. Radium-228 and radium-224 are decay products of thorium-232. Radium-226, a decay product of the uranium-238 decay chain (fig. 2), is the most abundant radium isotope in the environment (Szabo and others, 2012). Radium can be very soluble and chemically reacts similarly to other divalent alkaline-earth cations such as calcium, strontium, and barium; radium solubility can be enhanced by the common-ion effect where competing cations are abundant (Focazio and others, 2001). Once dissolved, radium-226 and radium-228 can be mobile in groundwater for considerable periods of time because of their relatively long half-lives (1,622 years (yr) and 5.75 yr, respectively) (Szabo and others, 2012). In contrast, radium-224 has a relatively short half-life (3.64 days).

Radon-222 is in the uranium-238 decay series and is the decay product of radium-226. Radon occurs naturally in water as a dissolved noble gas, and is virtually inert because it does not combine with other chemicals. With a half-life of about 3.8 days, radon-222 in groundwater does not travel far from its source. Radon concentrations can be elevated in groundwater, but the gas is volatile and dissipates quickly when water is exposed to the atmosphere. Therefore, radon concentrations typically are low in surface water but can be elevated in indoor air as the groundwater is used in the home and volatilizes. In fact, aeration is a treatment option to reduce radon levels in domestic well water (U.S. Environmental Protection Agency, 2012a).

Polonium-210 is the predominant isotope of polonium and is widely distributed in small concentrations in the Earth's crust. Polonium-210 is the last radioactive decay product in the uranium-238 decay series and has a half-life of 138 days. Polonium-210 is a highly toxic alpha emitter and is rarely found in groundwater at activities exceeding 1 picocurie per liter (pCi/L) (Seiler, 2011).
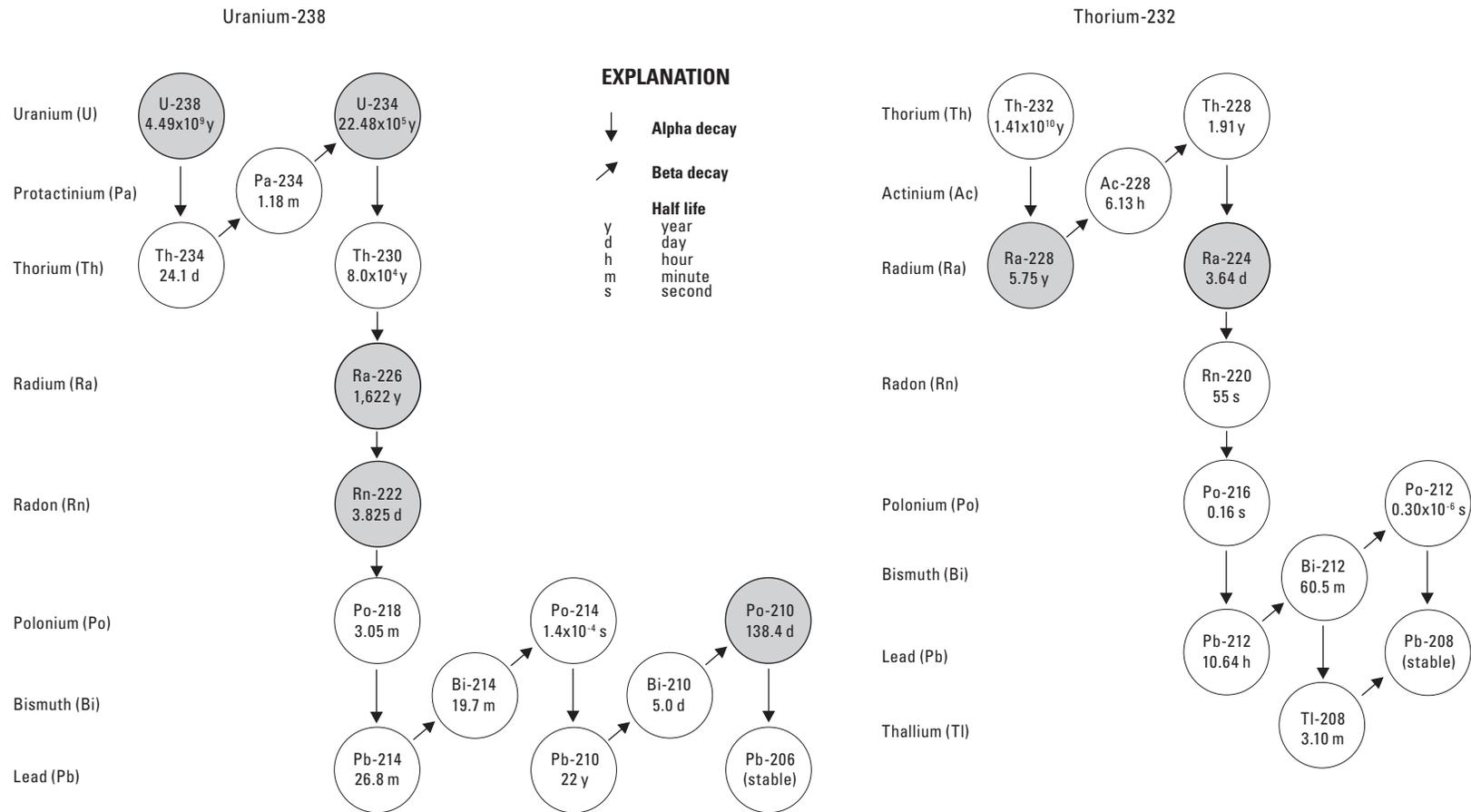
Figure 2. Uranium-238 and thorium-232 radioactive decay series (radionuclides analyzed for this study are shaded; uranium-235 and gross alpha-particle and gross beta-particle activity also were analyzed for this study but are not shown; modified from Oden and others, 2011).

The overall radioactivity of a water sample can be quantified by measuring the gross alpha- and beta-particle activities. Gross alpha-particle activity is a measure of the total amount of radioactivity attributable to the radioactive decay of alpha-emitting isotopes such as uranium-238 and radium-226. Alpha radiation is composed of a particle, consisting of two protons and two neutrons, that is spontaneously emitted from the nucleus of an alpha-emitting isotope of a radioactive element during radioactive decay. Beta-particle radiation, measured as gross beta-particle activity, has the composition of a particle, consisting of an electron that is spontaneously emitted from the nucleus of a beta-emitting isotope of a radioactive element, such as potassium-40 and radium-228, during radioactive decay. Emissions of both alpha and beta particles are considered ionizing radiation because they are energetic enough to break chemical bonds, thereby possessing the ability to damage or destroy living cells (Sloto, 2000).

Detailed overviews of interpreting and reporting radiological water-quality data are included in McCurdy and others (2008) and Oden and others (2011). The USGS currently (2013) reports the gross activity of a radionuclide in curies (Ci). One curie equals 3.7×10^{10} becquerels (Bq), the International System (SI) unit of radioactivity. Radionuclide concentrations and activities are commonly reported in pCi/L, which equals 2.22 radioactive disintegrations per minute (0.037 disintegrations per second) per liter of water. Picocuries (pCi = 10^{-12} Ci) per liter is a unit expressing the concentration (activity) of radionuclides in solution as particles emitted per unit volume (liter) of water (Focazio and others, 2001). By definition, 1 gram of radium has 1 curie of activity.

The activity of a single radionuclide (for example, radium-226) is considered a concentration because mass is linked directly to activity by the radioactive decay equation and half-life. Gross activity measurements (for example gross alpha-particle activity) are considered activities and not concentrations because there is no identifiable half-life or isotope mass associated with the measurement.

Drinking-Water Standards and Potential Health Effects

Exposure to elevated concentrations of radioactive constituents is a human-health concern because they are human carcinogens if ingested or inhaled. Ionizing radiation can cause cellular tissue damage, which in turn can increase the risk of incurring cancer (U.S. Environmental Protection Agency, 1999a). A maximum contaminant level (MCL) is the highest level of a contaminant that is allowed in drinking water. MCLs are legally enforceable U.S. Environmental Protection Agency (USEPA) drinking-water standards for public water-supply systems that deliver service to at least 15 connections and to at least 25 individuals. These regulations are not enforceable for domestic wells. The USEPA sets a MCL as close as possible to a maximum contaminant level goal (MCLG), which is level of a contaminant in drinking water below which there is no known or expected health risk (U.S. Environmental Protection Agency, 2000). The USEPA regulations for radioactive constituents in drinking-water were updated in December 2000 (U.S. Environmental Protection Agency, 2000). Current (2013) USEPA regulations for radioactive constituents in drinking-water are listed in table 1 (U.S. Environmental Protection Agency, 2009). State of Montana regulations (Montana Department of Environmental Quality, 2012) for radionuclides in drinking water are equal to those of USEPA, with the exception of radon (see below). USEPA allows for holding times of 6 months before analysis for analysis of gross alpha-particle activity, combined radium-226 and radium-228, and uranium. Radionuclide samples for this study typically were analyzed within a few days after collection, and therefore data for this study may more closely represent the concentrations and activities of water being consumed locally from domestic wells.

The MCL for uranium is 30 micrograms per liter (µg/L) (U.S. Environmental Protection Agency, 2009). Exposure to uranium in drinking water can result in chemical and radiological toxicity, including toxic effects to the kidney (U.S. Environmental Protection Agency, 2009). Some people who drink water containing alpha emitters, such as uranium, in excess of the MCL over many years may have an increased risk of developing cancer (U.S. Environmental Protection Agency, 2012b).

Table 1.   U.S. Environmental Protection Agency maximum contaminant levels for radiochemical constituents in drinking water, 2013.

[pCi/L, picocuries per liter; mrem/yr, millirems per year; µg/L, micrograms per liter; MMM, Multimedia Mitigation; MCL, maximum contaminant level]

Constituent	Maximum contaminant level
Uranium (natural)	30 µg/L[1]
Radon-222	300 or 4,000 pCi/L[2]
Gross alpha-particle activity	15 pCi/L[1,3]
Beta / photon emitters	4 mrem/yr (50 pCi/L screen)[1,4]
Combined radium (radium-226 plus radium-228)	5 pCi/L[1]

[1]Drinking Water Contaminants, National Primary Drinking Water Regulations (U.S. Environmental Protection Agency , 2009).

[2]Proposed U.S. Environmental Protection Agency maximum contaminant level (1999c) for states that choose to develop enhanced state programs to address the health risks from radon in indoor air—known as Multimedia Mitigation (MMM) programs. If a state chooses not to develop an MMM program, individual water systems in that state would be required to either reduce radon in their system for drinking water to the proposed maximum contaminant level (MCL) of 300 pCi/L or develop individual local MMM programs and reduce levels in drinking water to the alternative MCL of 4,000 pCi/L.

[3]Excluding radon and uranium.

[4]Dose in millirems per year determined on basis of individual isotope analyses. Gross beta-particle activity of 50 pCi/L is considered a screening level for further evaluation (California Department of Public Health, 2008).

The USEPA proposed a regulation for radon-222 in drinking water in 1999, with a MCL of 300 pCi/L (U.S. Environmental Protection Agency, 1999c), that has yet (2013) to be adopted. The proposed radionuclide rule also defined an alternative radon-222 MCL of 4,000 pCi/L. The proposed radon-222 standard of 4,000 pCi/L would be used to regulate community water systems with established multimedia programs to address radon-222 risks. To date (2013), these proposed radon-222 MCLs are considered guidance levels defining a range of maximum acceptable exposure risks. The State of Montana water-quality standards (Montana Department of Environmental Quality, 2012) lists a human health standard of 300 pCi/L for radon in groundwater.

Alpha particles are highly ionizing, but the particles travel short distances in air (less than 2 inches) before being absorbed (Sloto, 2000). Alpha particles move slowly and cannot penetrate skin (Szabo and others, 2005). However, radionuclides that are dissolved in water and ingested can emit alpha- and beta-particle radiation that can come into contact with, ionize, and damage internal cell tissue (Szabo and others, 2005). The USEPA MCL is specified in terms of adjusted gross alpha-particle activity and is listed as 15 pCi/L; the gross alpha-particle activity is "adjusted" by subtracting the activity caused by uranium and radon from the gross alpha-particle activity value (U.S. Environmental Protection Agency, 2000). The USEPA also specifies maximum allowable holding times before analysis, 6 months for individual samples or 12 months for composite samples (U.S. Environmental Protection Agency, 2000).

Alpha-particle activities from this study are listed in this report as (1) actual lab-reported gross alpha-particle activities and (2) adjusted gross alpha-particle activities. Radon activity was effectively removed from the sample during the analytical process. The adjusted gross alpha-particle activities were calculated by subtracting measured or estimated uranium values from the laboratory-reported alpha-particle activities. Samples from this study were analyzed within a shorter time period than specified in USEPA guidelines for public water-supply systems. Samples were analyzed at two different times (at approximately 72 hours and about 30 days after sample collection); these analyses are referred to as "72-hour count" and "30-day count" in this report. Laboratory-reported gross-alpha-particle activities and the short time period between sample collection and analysis likely more closely represent alpha-particle activities consumed in water from a domestic well.

Beta particles usually travel greater distances in air (about 6 feet) than alpha particles before being absorbed (Sloto, 2000). Beta particles can present an external as well as an internal hazard from inhalation or absorption, as they can penetrate the surficial layer of skin (Szabo and others, 2005). The U.S. Environmental Protection Agency (2000) gross beta-particle activity MCL is 4 millirems per year (mrem/yr) as it applies to annual dose equivalent to the total body or any internal organ from man-made beta-particle and photon emitters. The units of picocuries per liter cannot be directly converted

to millirem without identification and quantification of radionuclides in the sample. However, elevated levels of gross beta-particle activity may be used to screen for presence of beta-emitting radionuclides that warrant further investigation. The California Department of Public Health drinking water standard (CA-MCL) is 4 mrem/yr and 50 pCi/L (California Department of Public Health, 2008). A gross beta-particle activity of 50 pCi/L is set as a screening level for this report. Samples from this study were analyzed for gross beta-particle activity twice, at approximately 72 hours (72-hour count) and about 30 days (30-day count) after sample collection.

The radium MCL is defined as 5 pCi/L for the sum of the radium isotopes, radium-226 and radium-228 (U.S. Environmental Protection Agency, 2009). Radium has similar characteristics to calcium in the human body and can replace calcium in bones (Szabo and others, 2012). Currently (2013), there is no MCL for radium-224. However, Parsa (1998) calculated that radium-224 could contribute considerable gross alpha-particle activity to a water sample for about 7 days after sample collection. Although the USEPA has yet to establish a drinking-water standard for radium-224, the State of New Jersey has implemented a maximum 48-hour holding time before water samples are analyzed to include the contribution of radium-224 decay (New Jersey Department of Environmental Protection, 2002). As an alpha emitter, contributions from radium-224 are indirectly assessed with the USEPA MCL for adjusted gross alpha-particle activity of 15 pCi/L (U.S. Environmental Protection Agency, 2000).

Polonium-210 currently (2013) does not have an established MCL, but is considered to be a radionuclide of potential human-health concern and was analyzed in water samples for this study. Elevated concentrations of polonium-210 can be of concern as a contributor to gross alpha-particle activity, as indicated in a study of groundwater in Nevada (Seiler, 2011). Polonium-210 has been proposed for inclusion on the list of contaminants under the Unregulated Contaminant Monitoring Rule (U.S. Environmental Protection Agency, 1999b). As an alpha emitter, contributions from polonium-210 can be indirectly assessed with the USEPA MCL for adjusted gross alpha-particle activity of 15 pCi/L (U.S. Environmental Protection Agency, 2000).

Description of Study Area

The study area encompasses nearly 7,800 square miles (mi^2) in southwestern Montana and includes Jefferson County as well as adjacent parts of Lewis and Clark, Silver Bow, Deer Lodge, Powell, Broadwater, and Madison Counties (fig. 1). Although a small part of Gallatin County occurs within the study area boundaries, no groundwater quality data from that area were included in this study. Nearly one-half of the study area (49 percent) consists of public lands managed by local, State, and Federal government agencies. These public lands contain few wells, therefore most available wells are found in the more densely populated areas on private lands.

Hydrogeologic Framework

Geologic units in the study area range in age from Quaternary through Precambrian and include unconsolidated to consolidated sedimentary deposits, volcanic rocks, and igneous intrusive rocks. Geologic units within the study area were generalized for illustrative purposes to compare the water quality of groundwater within various rock types. Generalized geologic units for this study and corresponding geologic units of Ross and others (1955) are described in table 2 and shown on figure 3. Each of these geologic units is capable of yielding water to wells or springs and therefore is used as a source of groundwater. Wells completed in these units typically produce enough water for domestic or stock purposes, but locally some wells produce higher volumes suitable for other uses such as public water-supply systems. Before this investigation, some public water-supply wells have been determined to produce water with elevated uranium concentrations at or above drinking-water standards (Montana Department of Environmental, 2013).

Unconsolidated Quaternary sediments (Qs) and unconsolidated to consolidated Tertiary sediments (Ts) are found locally as basin-fill deposits including alluvium, alluvial fan, alpine glaciation, volcanic ash, and erosional material from local bedrock sources. These sedimentary materials range from clay to coarse-grained deposits and are a primary source of groundwater. Tertiary sedimentary rocks located in the Helena and Townsend areas have been found to have high uranium and thorium concentrations (Becraft, 1955; Becraft, 1956b; Wopat and others, 1977).

The bedrock units including Tertiary through Cretaceous igneous intrusive rocks (TKi), Tertiary through Cretaceous volcanic rocks (TKv), Cretaceous through Cambrian undifferentiated rocks (KЄu), and Precambrian undifferentiated rocks (pЄu) are exposed in upland areas and occur at depth beneath the basin-fill deposits. Wells completed in the bedrock units typically produce a limited amount of water sufficient for domestic or stock purposes, although a limited number of wells do produce water at higher rates.

Tertiary through Cretaceous igneous intrusive rocks (TKi) primarily consisting of rocks of the Late Cretaceous Boulder batholith are widespread in the study area (Knopf, 1957; Smedes and others, 1988; du Bray and others, 2012). The entire extent of the Boulder batholith, a large mass of primarily granitic rock that cooled and crystallized at depth, is included in the study area (fig. 3). The Boulder batholith consists of the Butte Granite and a group of associated smaller intrusions into Mesoproterozoic to Mesozoic sedimentary rocks and into the Late Cretaceous Elkhorn Mountains Volcanics (du Bray and others, 2012). These granodiorite, monzogranite, and minor syenogranite intrusions contain varying abundances of plagioclase, alkali feldspar, quartz, biotite, hornblende, rare clinopyroxene, and opaque oxide minerals (du Bray and others, 2012).

Uranium and thorium concentrations are elevated in the Boulder batholith, particularly in the abundant silicic granitic rock types (Becraft, 1956a; Becraft, 1956b; Jarrard, 1957; and Tilling and Gottfried, 1969). Castor and Robins (1978) reported that areas near Boulder, Butte, and Clancy may be favorable for large, low-grade uranium deposits. Specifically, uranium-bearing minerals reported in the rocks of the Boulder batholith and similar units in the area include the following: pitchblende (commonly composed of UO_2); uraninite (UO_2); torbernite ($Cu(UO_2)_2(PO_4)_2 \cdot 12H_2O$; zeunerite ($Cu(UO_2)_2(AsO_4)_2 \cdot 12H_2O$; autunite ($Ca(UO_2)_2(PO_4)_2 \cdot 11H_2O$; metatorbernite $Cu(UO_2)_2(PO_4)_2 \cdot 8H_2O$; uranocircite ($Ba(UO_2)_2(PO_4)_2 \cdot 10H_2O$; rutherfordine (($UO_2)CO_3$); uranophane ($Ca(UO_2)_2(HSiO_4)_2 \cdot 5H_2O$; and voglite ($CaCu(UO_2)(CO_3)_4 \cdot 6H_2O$) (Roberts and Gude, 1953a,b).

Tertiary through Cretaceous volcanic rocks (TKv) are found in close proximity to the Boulder batholith. Elkhorn Mountains Volcanics and the Boulder batholith are part of the same magmatic system (Smedes, 1966). The Elkhorn Mountains Volcanics represent magma that reached the surface and erupted, forming a volcanic and volcanogenic layered sequence; the Boulder batholith represents the parent magma that crystallized beneath the Elkhorn Mountain Volcanics (Robinson and others, 1968; Rutland and others, 1989). Younger Eocene and Oligocene volcanic rocks including the Lowland Creek Volcanics and associated dikes are not related to the Boulder batholith but occurred later as regionally important igneous events (O'Neill and others, 2004). Uranium and thorium concentrations are elevated in rhyolitic volcanic rocks overlying the Boulder batholith (Tilling and Gottfried, 1969).

The generalized Cretaceous through Cambrian undifferentiated sedimentary rock (KЄu) unit consists of a wide variety of geologic units (table 2). This generalized unit includes sedimentary rocks of variable depositional history, sources, and mineralogy. Specifically, only three wells sampled for this study were completed in this generalized unit including sandstone of the Colorado Group of Cretaceous age, limestone of the Madison Group of Mississippian age, and limestone of Devonian age (Jefferson Formation as reported in Thamke and Reynolds, 2000).

The oldest strata in the study area are generalized as the Precambrian, undifferentiated rocks (pЄu) that primarily include rocks of the Belt Supergroup (called Belt series in Ross and others, 1955), which are a thick sequence of typically fine-grained siliciclastic and carbonate strata exposed in northern and southern parts of the study area (Ross and others, 1992). This group of sedimentary and metamorphic rocks as much as 18 kilometers thick was deposited approximately 1,400 million years ago on a crystalline basin extending southward from Canada, through western Montana, into northern and central Idaho, and into eastern Washington. Within the study area, several units within the Belt Supergroup are exposed or near land surface including the Helena, Spokane, and Greyson Formations (table 2).

Table 2.   Generalized geologic units in Jefferson County and surrounding areas, Montana.

System	Generalized geologic unit	Geologic unit (s) modified from Ross and others (1955)[1]	Hydrogeologic unit(s) of study wells[2]	Description
Quaternary	Qs—Quaternary sediment	Alluvium (Qal), glacial drift (Qg), and terrace deposits (QTt)	Quaternary alluvium	Variable deposits of unconsolidated gravel, sand, silt, and clay.
Tertiary	Ts—Tertiary sediment	Tertiary sedimenary rocks, undifferentiated (Ts)	Tertiary sediments or Climbing Arrow Formation	Unconsolidated sediments including clay, silt, sand, and gravel and moderately consolidated siltsone, sandstone, and conglomerate.
Tertiary through Cretaceous	TKi—Tertiary through Cretaceous igneous intrusive rocks	Tertiary dikes and sills (Td), Tertiary coarse-grained intrusive rocks (Tg), Tertiary coarse-grained intrusive rocks (Tga), Boulder batholith and broadly related rocks (TKb), Diorite and gabbro (Kdg)	Boulder batholith intrusives, Marysville intrusives, or upper Cretaceous plutonic rocks	Primarily intrusive rocks of the Late Cretaceous Boulder batholith and other igneous intrusive rocks.
Tertiary through Cretaceous	TKv—Tertiary through Cretaceous volcanic rocks	Tertiary volcanic rocks (Tv) and Cretaceous volcanic rocks (Kv)	Tertiary volcanic rocks or Elkhorn Mountain Volcanics	Primarily volcanic rocks of the Lowland Creek Volcanics and the Elkhorn Mountains Volcanics (U.S. Geological Survey, 2012a).
Cretaceous through Cambrian	K€u—Cretaceous through Cambrian undifferentiated rocks	Two Medicine Formation (Ktm); Colorado Group (Kc); Kootenai Formation and associated rocks (Kk); Jurassic, undifferentiated (Ju); Triassic, undifferentiated (Tru); Permian, undifferentiated (Pu); [1]Mississippian, undifferentiated (Mu, Madison Group in U.S. Geological Survey, 2012a; Devonian, undifferentiated (Du); and Cambrian undifferentiated (€u)	Jefferson Limestone, Dolomite, Formation, or Group, the Colorado Group, or the Madison Group	A variety of sedimentary bedrock units.
Precambrian	p€u—Precambrian undifferentiated rocks	Missoula Group (p€m), [1]North Boulder Group (P€nb, LaHood Formation in U.S. Geological Survey, 2012a), [1]Helena Dolomite (p€h, Helena Formation in U.S. Geological Survey, 2012a), [1]Spokane shale (p€s, Spokane Formation in U.S. Geological Survey, 2012a), Greyson Formation (p€g), Newland Limestone (p€n), Ravalli Group (p€r), and Pre-Belt Supergroup gneiss, schist, and related rocks (p€gs)	Spokane shale, Belt Group, Helena Dolomite, Greyson Formation, or Mount Shields Formation	Primarily a variety of sedimentary bedrock units within the Belt Supergroup.

[1]Geologic units modified from Ross and others (1955) with nomenclature from U.S. Geological Survey National Geologic Map Database (2012a).

[2]The U.S. Geological Survey National Water Information System database hydrogeologic unit(s) assigned to the open interval of study wells located within this generalized geologic unit (appendix 2).

EXPLANATION

Generalized geologic unit—See table 2 for descriptions

- Qs   Quaternary sediment
- Ts   Tertiary sediment
- TKi   Tertiary through Cretaceous igneous intrusive rocks, includes rocks of the Boulder batholith
- TKv   Tertiary through Cretaceous volcanic rocks, includes rocks of the Lowland Creek Volcanics and the Elkhorn Mountain Volcanics
- KЄu   Cretaceous through Cambrian undifferentiated rocks
- pЄu   Precambrian undifferentiated rocks

Base modified from U.S. Census Bureau TIGER digital data, 1993–2001,1:100,000
Universal Transverse Mercator Projection, Zone 12
North American Datum of 1927 (NAD 27)

Geology modified from Ross and others (1955)

Figure 3.   Generalized geology of Jefferson County and surrounding areas, Montana.

Water moves through the groundwater flow system either through spaces (pores) between individual particles within the sedimentary aquifers (such as Qs, Ts, and KЄu) or through fractures within crystalline rocks (such as TKi and TKv) or some sedimentary to metamorphic rocks (such as pЄu) (fig. 4).

Recharge to the groundwater flow system occurs as infiltration from precipitation, leakage from streams and canals, and as leakage from underlying rocks. Discharge from the groundwater system occurs by withdrawal from wells, seepage to streams and drains, and evapotranspiration (ET) (Kendy and Tresch, 1996).

NOT TO SCALE

Groundwater flow through pores in sedimentary aquifers

Groundwater flow through fractures in crystalline rocks

EXPLANATION

Alluvium—Unconsolidated sand and gravel

Sedimentary rock—Consolidated sediments

Crystalline rock —Including granitic and volcanic rocks

Generalized groundwater flow

Well

Figure 4.   A generalized groundwater flow system.

Groundwater quality is affected naturally by (1) the mineralogy of aquifer material, (2) geochemical reactions associated with the physical properties of the water (such as pH, temperature, and dissolved oxygen), and (3) residence time (the length of time water is in contact with source minerals). Groundwater velocities through porous media or fractures is highly variable, thus the residence time for geochemical reactions such as mineral precipitation and dissolution, ion exchange, sorption, and radioactivity decay also will vary. Moreover, the diverse chemical makeup of the groundwater and the aquifer material can cause variations in groundwater quality. This variation can occur even in water produced from closely spaced wells because the sampled groundwaters may have traveled along different flow paths (fig. 4), and encountered varied subsurface mineralogy and geochemical processes.

Methods

A regional network of 128 wells was sampled to assess the distribution of concentrations of radioactive constituents including radionuclides, gross alpha-particle activity, and gross beta-particle activity in groundwater of Jefferson County and surrounding areas of southwestern Montana. Water samples from all 128 wells wer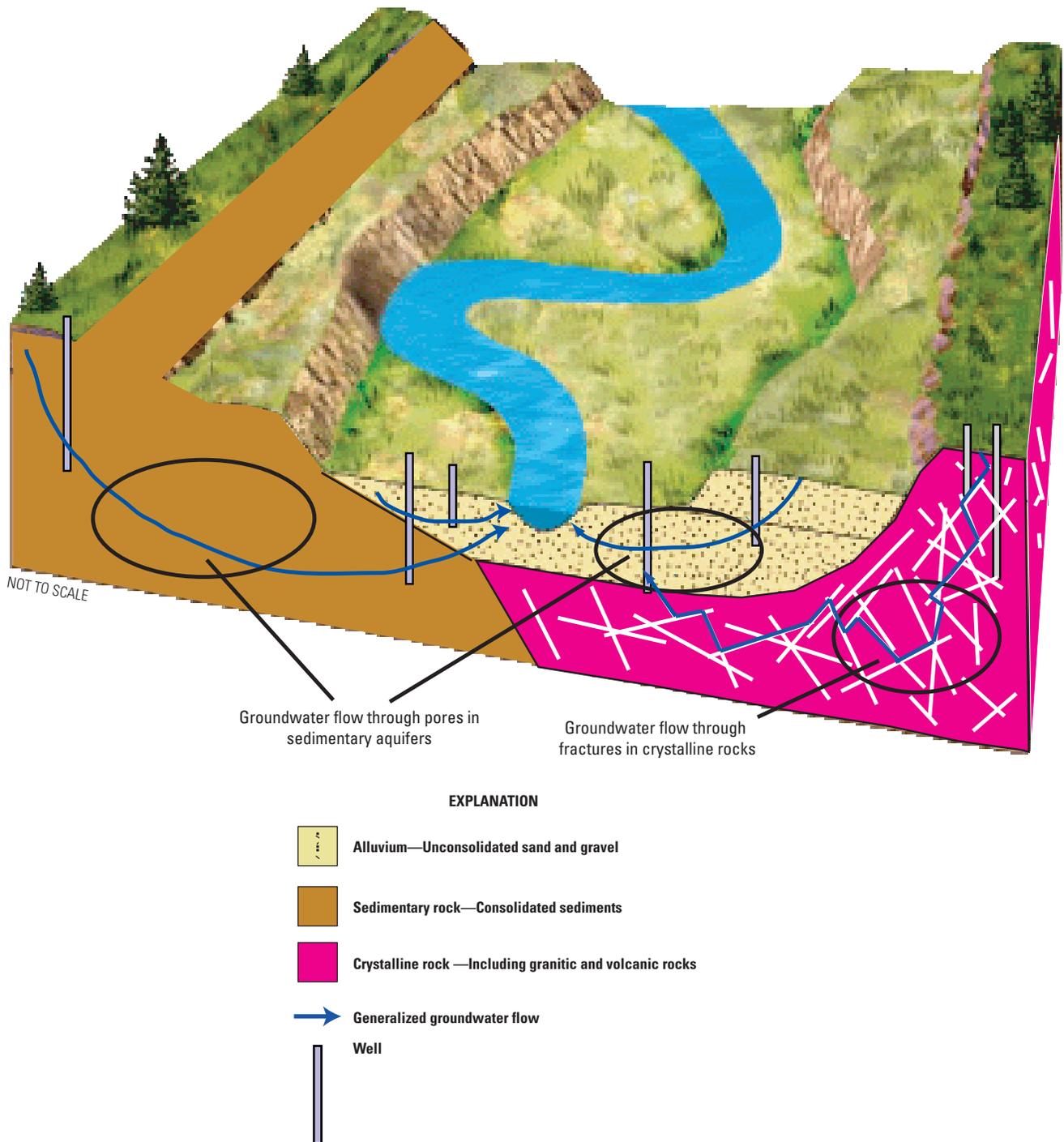e analyzed for dissolved uranium. Water samples also were analyzed for radon (127 wells), gross alpha- and beta-particle activity (104 wells), uranium and radium isotopes (46 wells), and polonium-210 (39 wells). Forty wells were sampled in 2007 and another 88 wells were sampled in 2009 and 2010. In addition, 39 of the 128 wells that had elevated radioactive constituents in the first sample were sampled a second time for additional analytes and to examine temporal variability.

The occurrence and concentrations of radiochemical constituents analyzed from groundwater samples collected as part of this study are described in relation to drinking-water standards and were examined spatially and by geologic unit. Potential relations between radioactive constituents in groundwater and geologic units as well as radioactive constituents and other chemical properties such as pH, dissolved oxygen, dissolved constituents, and calculated reduction/oxidation (redox) conditions were examined. Redox conditions of sampled waters were determined based on available dissolved oxygen, nitrate (NO_3^-), manganese (Mn^{2+}), iron (Fe^{2+}), and sulfate (SO_4^{2-}) data using a digital software program (Jurgens and others, 2009) based on the framework created by McMahon and Chapelle (2008).

In addition to data collected for this study, existing uranium and radon water-quality data for wells within the study area were retrieved from the USGS National Water Information System (NWIS) database (accessed on June 25, 2012, at *http://nwis. waterdata.usgs.gov/mt/nwis/qwdata*) and from the Montana Bureau of Mines and Geology (MBMG) Groundwater Information Center (GWIC) database (accessed on February 7, 2012, at *http://mbmggwic.mtech.edu/*). Only wells with uses listed as domestic, irrigation, stock, and commercial were included in this

retrieval. The water-sampling and analytical methods used to generate the dataset retrieved from GWIC may differ from those used by the USGS. The combined USGS and MBMG GWIC datasets were used to evaluate the general occurrence and concentrations of uranium and radon within the study area.

Radiochemical data also were compared to a national dataset collected for the USGS National Water Quality Assessment (NAWQA) program. Radiochemical data collected between 1992 and 2011 by the NAWQA program were retrieved from the USGS NAWQA Data Warehouse (U.S. Geological Survey, 2012b) and compared to data collected for this study. The NAWQA dataset is considered comparable to water-quality data collected for this study as the same USGS sampling procedures and analytical methods were followed. The NAWQA program includes regional groundwater assessments in 51 study areas (river basins and aquifers), referred to as Study Units, throughout the United States. The NAWQA study units include 19 regional aquifer systems that account for about 75 percent of the estimated groundwater withdrawals for drinking water for the Nation. Radiochemical data from this study also were compared to results reported in Ayotte and others (2011) and Szabo and others (2012) which included data collected as part of the NAWQA program.

Sampling Network

Wells included in this study span seven counties in southwestern Montana including Jefferson, Lewis and Clark, Silver Bow, Deer Lodge, Powell, Broadwater, and Madison0 Each well was assigned a unique site identification number and station name as described in appendix 1. Records of wells sampled for this study are included in appendix 2. Sampled wells included 1 public water-supply well, 1 monitoring well, 3 stock wells, and 123 wells used for the domestic supply of single households. Nearly one-half of the sampled wells (62 of 128) produced water from the Boulder batholith and other Tertiary through Cretaceous intrusive rocks (TKi), commonly described as granite. Additional wells were selected from geologic units commonly used for water supply in the area, including: alluvial sand and gravel deposits of the Quaternary sediments (Qs, 18 wells); consolidated to semi-consolidated Tertiary sedimentary rocks (Ts, 24 wells) including the Climbing Arrow Formation; Tertiary to Cretaceous volcanic rocks (TKv, 12 wells) including the Elkhorn Mountains Volcanics and the Lowland Creek Volcanics; Cretaceous through Cambrian undifferentiated rocks (KЄu, 3 wells) including wells in limestone of the Jefferson Formation and Madison Group and sandstone of the Colorado Group; and Precambrian undifferentiated rocks (pЄu, 9 wells) including shale, limestone, sandstone, and granite of the Belt Supergroup (appendix 2).

Most wells used for this study were randomly selected and lacked previous radiochemistry analyses. However, some were targeted (1) to evaluate occurrence of elevated radionuclide values of nearby wells, (2) at the special request of area residents, and (3) to examine the radiochemistry of five

close-proximity wells completed at variable depths in the Boulder batholith (delineated as targeted wells in appendix 2).

Random wells were selected using digital maps to generate a random sampling grid based on surficial geology with each generalized geologic unit divided into equal area cells (Scott, 1990). These maps then served as a guide for searching the MBMG GWIC database for existing water wells within each sampling grid. Existing wells were identified as candidate or potential sampling points based on pre-defined criteria intended to minimize the effects of well design on geochemistry. The candidate wells were selected based on discrete completion within the desired geologic units. Most sampled wells had existing submersible pumps or were sampled with a portable submersible pump if no pump was in place. The sampling density was not uniformly distributed over the study area because large tracts of the study area (49 percent) consist of publicly owned land with only a few existing wells available for sampling. Study wells were overwhelming located in more-populated, privately owned lands, which typically are in the lowlands (fig. 1).

Candidate wells were evaluated in the field to (1) gain permission from well owners, (2) ensure accuracy of GWIC well log descriptions, and (3) ensure the sampling point was not affected by treatment (filters, softeners) or storage systems. Each reconnaissance visit included the visual inspection of the sampling point (hydrant, faucet, and so on) and location of the well using a handheld global positioning system (GPS) unit. These wells were established in the U.S. Geological Survey National Water Information System database according to standard USGS procedures (Cunningham and Schalk, 2011) with unique a unique identifier described in appendix 1.

Sample Collection and Analysis

Groundwater samples were collected from 2007 through 2010 and processed using "ultra-clean" field techniques in accordance with standard USGS protocols (USGS National Field Manual for the collection of water-quality data, U.S. Geological Survey, variously dated). These procedures ensure a representative sample is collected at each site, and samples are collected and processed to minimize the potential for sample contamination. Samples were collected as near to the well head as possible and prior to treatment, generally from frost-free hydrants.

Field properties were measured with a multiparameter water-quality sonde equipped with an air-tight flow-thru chamber. The multiparameter sonde was calibrated prior to sample collection following procedures outlined in the USGS National Field Manual (U.S. Geological Survey, variously dated). Temperature accuracy was checked against a laboratory calibration thermometer in the laboratory prior to the onset of sampling. Specific conductance, pH, and dissolved oxygen were calibrated at field sites prior to sampling. Specific conductance was calibrated with one standard and then verified with another standard to check instrument calibration over a range of two standards. pH was calibrated using two

standards (pH 7 buffer and either pH 4 or 10 buffer). Dissolved oxygen was calibrated with a 1-point calibration at 100 percent saturation.

Prior to sampling, each well was purged until water-quality properties (pH, specific conductance, temperature, and dissolved oxygen) stabilized or until at least one bore volume of water was removed in recently used domestic or public water-supply wells (assuming water is frequently removed from the well during normal use). Approximately three bore volumes were removed in less frequently used wells (for example, stock wells or monitoring wells). Samples were collected when three to five consecutive field-parameter readings (collected at 3 to 5 minute intervals) were within USGS stability criteria (U.S. Geological Survey, variously dated). During sample collection, the sampled water came into contact with just the well, existing pump and plumbing system or portable stainless steel submersible pump, and Teflon (high density) tubing with stainless steel connectors connected directly to the well hydrant, or in rare instances, to a spigot on the house. All field equipment was cleaned according to standard protocol (U.S. Geological Survey, variously dated) to minimize contamination between wells. All samples, with the exception of samples for radon-222 analyses, were filtered through 0.45-micron disposable polypropylene capsule filters and collected within a portable processing chamber. Filtered samples were titrated in the field to determine alkalinity. Samples requiring preservation were preserved immediately on collection with laboratory-grade, ultra-pure, nitric acid. Unfiltered samples for radon-222 analyses were collected outside the sampling chamber with a syringe, through a pressurized brass valve with a Teflon septum to minimize degassing and exposure to the atmosphere. Samples requiring analysis of time-sensitive analytes (such as radon, gross alpha-particle activity, gross beta-particle activity, radium, and nutrients) were shipped overnight to the appropriate laboratory.

Samples for this study were analyzed for radionuclides, general chemistry (major ions, barium, iron, manganese, vanadium, and nutrients) at the USGS National Water Quality Laboratory (NWQL) in Denver, Colorado, and Eberline Analytical Services in Richmond, California (table 3). The NWQL maintains National Environmental Laboratory Accreditation Conference (NELAC) and other certifications (U.S. Geological Survey, 2010). The NWQL staff monitored the performance of Eberline Analytical Services by evaluating analytical results for test samples as well as the reported analytical data for groundwater samples collected for this study. The results for each radiological constituent and sample matrix combination were evaluated for technical issues and compliance to specifications stated in the performance work statements for the commercial laboratory (McCurdy and others, 2008). The acceptable data were then transferred to the USGS NWIS database and provided to the USGS Montana Water Science Center staff for further analysis, interpretation, and publication. Data collected for this study are available through the USGS NWIS database accessible through the web at *http://nwis.waterdata.usgs.gov/mt/nwis/qwdata*.

Table 3. Summary of analytes, analytical methods, analyzing laboratories, and field processing for chemical constituents in groundwater samples collected in Jefferson County and surrounding areas, Montana, 2007 through 2010.

[U.S. Geological Survey analytical schedule numbers are used for a suite of constituents, whereas laboratory codes (lab codes) are used for individual constituents. NWQL, U.S. Geological Survey National Water Quality Laboratory, Denver, Colorado; μm, micrometer; HNO_3, nitric acid; Eberline, Eberline Analytical Services, Richmond, California; HASL, Health and Safety Laboratory; NAS-NS, National Academy of Sciences-Nuclear Science Series]

Constituent or constituent group	Analytical Method	Reference	Laboratory	Filtration	Preservation
Major ions, iron, and manganese (schedule 1248, lab codes 645 and 1793)[1]	Inductively coupled plasma-mass spectrometry	Fishman and Friedman (1989), Fishman (1993)	NWQL	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Uranium, natural (lab code 1797), barium and vanadium (lab codes 1786 and 3134)	Inductively coupled plasma-mass spectrometry	Garbarino and others (2006)	NWQL	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Nutrients (schedule 1975 and lab code 3117)	Various methods	Fishman (1993), Patton and Kryskalla (2003)	NWQL	0.45 μm filter	Chilled to 0 to 4 degrees Celsius.
Radon-222 (lab code 1369)	Liquid scintillation following American Society for Testing and Materials method D5072-98	American Society for Testing and Materials International (1998)	NWQL	none	Agitate in equal part mineral oil.
Radium-224, Radium-226 (lab code 2164)	Alpha spectrometry following following Eichrom Technologies method RAW03	Eichrom Technologies, Inc. (2005)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Radium-226 (lab code 794)	Radon emanation following U.S. Environmental Protection Agency method 903.1 (emanation)	U.S. Environmental Protection Agency (2008)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Radium-228 (lab code 1364)	Radiochemical separation and beta counting following U.S. Environmental Protection Agency method 904.0 with modifications	U.S. Environmental Protection Agency (2008)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Gross alpha- and gross beta-particle activity (lab code 2802)	Followed U.S. Environmental Protection Agency method 900.0 within 72 hours of sample collection and again 30 days after sample collection; gross alpha-particle activity used thorium-230 as the calibration standard and gross beta-particle activity used cesium-137 as the calibration standard	U.S. Environmental Protection Agency (2008)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Uranium 234, 235, 238 (schedule 1130)	Alpha spectrometry following American Society for Testing and Materials method D3972-97	American Society for Testing and Materials International 1997)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.
Polonium-210 (lab code 1505)	Alpha spectrometry following Health and Safety Laboratory HASL-300 or NAS-NS 3037	HASL-300 (U.S. Department of Energy, 1997) or NAS-NS 3037 (Figgins, 1961)	Eberline	0.45 μm filter	Acidify with HNO_3 to below pH 2.

[1]Major ions include calcium, chloride, fluoride, magnesium, potassium, silica, sodium, and sulfate.

Concentration and activity data for radionuclides analyzed for this study are reported as decay corrected, which means that reported activities were calculated back to the time of sample collection based on decay constants. For example, radon-222 concentrations are calculated and reported at the levels they would have been when the samples were collected. Applying this correction is especially important for short-lived radionuclides such as radon-222, polonium-210, and radium-224 because the decay-corrected values more adequately represents the levels of radionuclides produced for consumption from domestic wells. However, gross alpha- and beta-particle activities are not decay corrected but are reported as the activity at the time of analysis (counting). Unlike the individual radionuclide concentrations, gross alpha- beta-particle activities represent combined activities of all of the respective alpha-particle and beta-particle emitters, present in unknown quantities in a sample, and therefore time-dependent corrections cannot be made.

Gross alpha- and beta-particle activities were analyzed at 72 hours (allowing for the measurement of activities contributed from short-lived isotopes) and at 30 days after sample collection. The gross alpha- and beta-particle activities present in a sample are dependent on the specific period of time associated with the measurement (U.S. Environmental Protection Agency, 1997). Gross alpha-particle activity is reported in pCi/L as thorium-230. The measured radioactivity is compared with the activity of a thorium-230 calibration standard, and the results are reported in terms of equivalent quantities of the reference standard. Gross beta-particle activity is reported in picocuries per liter as cesium-137 (pCi/L as cesium-137). The measured radioactivity is compared with the activity of a cesium-137 calibration standard, and the results are reported in terms of equivalent quantities of the reference standard.

Reporting of Results

The analytical results of the samples collected for this study are described in this report through the use of summary tables, maps of occurrence and concentration, and graphs. Analytical results were compared and contrasted between geologic units, compared to USEPA drinking-water standards and health advisories (U.S. Environmental Protection Agency, 2000 and 2009) and in some instances, compared to results from other USGS studies (Ayotte and others, 2011; U.S. Geological Survey, 2012). The USEPA drinking-water standards include Maximum Contaminant Levels (MCLs), Alternative Maximum Contaminant Levels (AMCLs), and Secondary Drinking Water Regulations (SDWRs), and Lifetime Health Advisories (LHAs). MCLs are enforceable standards that apply to public water-supply systems and are defined as the highest allowable concentration of a contaminant in drinking water. An AMCL is an option to the MCL, but requires compliance with additional conditions (U.S. Environmental Protection Agency, 1999c). SDWRs are non-enforceable guidelines for constituents that may cause cosmetic or aesthetic effects in drinking water. LHAs are non-enforceable guidelines for noncarcinogens.

Several wells were sampled multiple times from 2007 through 2010 for this study. Summaries of ranges of concentrations included results of all analyses. Discussions of the number of wells that exceeded water-quality criteria included all analytical results and reported as wells that exceeded water-quality criteria in at least one sample. Median concentrations of samples from individual wells were used for (1) discussions/summaries of median concentrations of individual analytes and (2) discussions/summaries of median concentrations within individual geologic units.

Major ion and trace element water-chemistry data were reported by the NWQL relative to minimum reporting levels (MRLs) or laboratory reporting levels (LRLs). The MRL is the minimum concentration at which a constituent can be reliably reported using a given analytical method (Timme, 1995). The LRL is statistically calculated by evaluating quality-control data for an analytical method on a continuing basis to determine long-term method detection levels (LT-MDLs) (Childress and others, 1999). The NWQL reports analytical results as estimated values if the value is smaller than a minimum concentration reliably reported by a given analytical method, but larger than the long-term method detection level (Childress and others, 1999). Estimated values indicate that constituents have been identified in a sample, but the reported concentration has more uncertainty than concentrations not reported as estimated.

Radionuclide analytical results were reported by the laboratory as values rounded according to American National Standards Institute (2003) procedure N42.23 with associated 1-sigma combined standard uncertainty (CSU) and a sample-specific critical level (ssLc). Some results were reported as negative values, indicating that the radiation count of the sample was less than the long-term average background radiation routinely reported by the instrument (Troyer and others, 1991). The 1-sigma CSU is a calculated measure of uncertainty of the analysis and combines the laboratory and counting uncertainty. By definition, the true radionuclide concentration has a 68-percent probability of being within plus or minus the 1-sigma CSU of the reported value (McCurdy and others, 2008). An analyte is considered not detected (ND) when the concentration is less than the ssLc.

For each radionuclide measurement, the laboratory calculates a sample-specific minimum detectable concentration (ssMDC) that is computed individually for each radionuclide sample based on instrument operating conditions at the time of the measurement and background radioactivity over time (Focazio and others, 2001). The ssMDC is used by the laboratory to verify that the a priori minimum detectable concentration (MDC, defined as the lowest concentration resulting in a 95-percent confidence level that the value is greater than the ssLc (McCurdy and others, 2008)) was achieved.

Quality-Control Sample Collection and Data Analysis

Field blanks and replicates were collected as quality-control samples to identify potential sample contamination and examine analytical measurement variability and bias. Quality-control samples composed more than 10 percent of the samples collected for this study. Procedures for collecting quality-control samples are described in Koterba and others (1995). In addition to field samples, contract and USGS laboratories follow quality-assurance procedures that include evaluations of yields, recoveries, blank and duplicate results, and sample-specific parameters such as the critical level and minimum detectable concentration (Pritt and Raese, 1995; Maloney, 2005; McCurdy and others, 2008). The NWQL participates in a blind-sample program in which standard reference water samples prepared by the USGS Branch of Quality Systems are routinely inserted into the sample line for each analytical method at a frequency proportional to the sample load (*http://bqs.usgs.gov*). The NWQL also participates in external evaluation studies and audits with the National Environmental Laboratory Accreditation Program, the U.S. Environmental Protection Agency, Environment Canada, and the USGS Branch of Quality Systems, in order to assess analytical performance (Glodt and Pirkey, 1998) NWQL quality assurance results are available through the web at *http://wwwnwql. cr.usgs.gov/USGS_Quality.shtml*.

Field-blank sample results were used to determine if equipment decontamination procedures were adequate, and if sampling or laboratory analytical procedures resulted in sample contamination. Water certified as free of inorganic constituents was used for field-blank samples, which were collected immediately after environmental samples were collected and equipment was cleaned. Field blanks were analyzed for major ions, barium, iron, manganese, vanadium, nutrients, and radioactive constituents. Constituents detected in field blanks at concentrations less than 10 percent of the environmental concentrations did not result in significant bias in the environmental results.

Replicate samples consisted of two environmental samples considered identical in composition. In the field, two replicate sample bottles were filled sequentially, one immediately after the other. The combined effects of sample-collection variability and precision of analytical results were evaluated by calculating the relative percent difference (RPD) of the constituent concentrations in replicate analyses.

Variability for each analyte is estimated as the relative percent difference (RPD) between the two replicates (equation 1):

$$RPD = \frac{|C_{env} - C_{rep}|}{(C_{env} + C_{rep})/2} \times 100 \qquad (1)$$

where

$|C_{env} - C_{rep}|$ = absolute value of the difference between concentrations of the analyte in the primary environmental sample and the replicate sample, and

$(C_{env} + C_{rep})/2$ = mean concentration of the analyte in the primary environmental sample and replicate sample.

Field replicates were collected for major ions, barium, iron, manganese, vanadium, nutrients, and radioactive constituents. The RPD cannot be calculated if the concentration is censored in either or both samples. For this study, RPD values greater than 20 percent were considered indicative that analytical results might be affected by high variability. Analytes with RPDs outside this criterion are identified in the following data discussions.

Major-ion data were quality assured by calculating a cation-anion balance. The sum of concentrations of dissolved cations in milliequivalents per liter should equal the sum of concentrations of dissolved anions in milliequivalents per liter (Hem, 1985). The percent difference between the sum of concentrations of cations and anions in milliequivalents per liter was calculated using equation 2.

$$\text{Percent difference} = \frac{(\text{sum of dissolved cations - sum of dissolved anions})}{(\text{sum of dissolved cations+sum of dissolved anions})} \times 100 \quad (2)$$

Statistical Analysis

Summary statistics are reported in tables and graphically represented through the use of boxplots within this report. Summary statistics, which include the minimum, maximum, and median values were computed for all radiochemical constituents, major ions, and physical properties. Also included in the summary statistics are the range and median values of the laboratory reporting levels (LRLs) and sample specific critical levels (ssLcs) for each analyte. All results were included as retrieved from the USGS National Water Information System database when describing the range of values, although some of the lower end values are considered nondetects. Median values were calculated using all data, with the exception of individual wells sampled multiple times. The median values from individual wells sampled multiple times were used in the calculation of overall median values of constituents. Negative values were set equal to 0.001 (pCi/L or µg/L) for computational purposes. No other manipulation, censoring, or screening of the data was attempted to avoid biases resulting from the omission of low concentrations that might change the true statistical distribution of the radiochemistry dataset. Radiochemical results reported at values less than sample specific critical levels were included in statistical computations but were not included in the scatter plots or box plots in this report.

Associations between various radiochemical constituents and associations of radiochemical constituents with a variety of major-ion and physical properties were evaluated to examine the conditions in which elevated radionuclides exists. Associations were examined with all data collected for this

study and then associations were examined within the TKi geologic unit. All analytical results from wells sampled multiple times were included in this analysis because conditions may have varied between sampling events. Statistical tests and models were computed with the use of TIBCO Spotfire S+ 8.1 software (TIBCO Software Inc., 2008).

Nonparametric statistics, which do not require distributional assumptions about the censored data, were used to describe distributions or compare data among groups. Nonparametric methods offer several benefits, including no assumption of normally distributed data, good statistical power, and the use of data values that are less than the censoring level without having to create values for them, resulting in an accurate portrayal of the censored information (Helsel and Hirsch, 1992).

The nonparametric Spearman correlation coefficients (rho) were computed to measure the strength of the relation between radiochemical constituents and between radiochemical constituents and inorganic constituents and physical properties. Rho correlation coefficients can range from +1 to -1, which indicates a perfect positive association and a perfect negative association of ranks, respectively (Helsel and Hirsch, 1992). A rho near zero indicates a weak association between ranks. The null hypothesis for this test is that there is no association between the two variables in the dataset. The significance level (α) for rho statistical test was set equal to 0.05 for this study. Thus, for p-values less than 0.05, the null hypothesis can be rejected in favor of the alternate hypothesis. Therefore, a p-value of 0.05 indicates that there is a 5-percent chance that the strength of the relation (rho coefficient) happened by chance if the null hypothesis were true. For the purposes of this report, the terms used to describe statistically significant correlations are strong (greater than or equal to +0.9 to +1.00 or less than or equal to -0.90 to -1.00), moderate (from +0.50 to +0.89 or from -0.50 to -0.89), and weakly or not correlating (from -0.49 to +0.49). Correlations are evaluated as to the degrees of significance (p) and reported with the correlation value.

Spatial analysis of the available data is somewhat difficult because the wells were not entirely randomly distributed (some of the sites were targeted). In addition, the site-selection process for the wells with data retrieved from the GWIC database is not known. A nonparametric statistical test, the Wilcoxon rank-sum test (Helsel and Hirsch, 1992), was used to determine if water-quality differences existed between samples collected in specific areas (for example, near Clancy) and the remainder of the study area. The null hypothesis in each test was that there is no significant difference between the two groups of water-quality data that are grouped by location (for example: uranium concentrations in sampled Boulder batholith wells within 5 miles of Clancy as compared to uranium concentrations in sampled Boulder batholith wells elsewhere in the study area). The significance level (α) for the Wilcoxon rank-sum test was set equal to 0.05. For these tests, a significant difference in the values (that is, uranium) between sample groups was assumed if the p-value is less than or equal to 0.05.

General Chemistry

Physical properties and major-ion chemistry of samples collected from the 128 wells included in this study are tabulated in appendix 3 and summarized in table 4. The chemical composition of groundwater is dependent upon the original source of the water as well as geochemical reactions, such as dissolution of minerals, that occur as the water flows through the porous media, fractures in bedrock, or dissolution features. Depending on the mineralogy of the aquifer material and the physical properties of the groundwater (for example, pH, dissolved oxygen, and general water chemistry), radionuclides may be entrained or removed as groundwater moves along its flow path.

The concentrations of most nonradioactive constituents in the sampled waters did not exceed USEPA drinking-water standards (U.S. Environmental Protection Agency, 2000). However, nonradioactive constituents in some samples did have concentrations that exceeded USEPA MCLs and SDWRs (table 4). Although MCLs are set to minimize human-health hazards, SDWRs were established as guidelines to assist public water-supply systems in managing their drinking water for cosmetic, aesthetic, or taste considerations. U.S. Environmental Protection Agency MCLs were exceeded for fluoride (2 of 109 wells) and nitrate (5 of 127 wells). The USEPA SDWRs of pH (above or below the recommended pH range in 8 of 128 wells), total dissolved solids (21 of 128 wells), chloride (1 of 128 wells), fluoride (4 of 109 wells), sulfate (9 of 128 wells), iron (12 of 128 wells), and manganese (22 of 98 wells) were exceeded. The major-ion chemistry of the sampled waters varied, however most (about 70 percent) are classified as calcium-bicarbonate type waters.

Quality Assurance Results of Major Ions

Nine sets of replicate samples were analyzed for major ions to assess variability in results caused by sample-collection procedures and laboratory analysis. Replicate sample results and associated relative percent differences are listed in appendix 4. Analyses of the replicate samples indicated good agreement between the measured concentrations. The relative percent differences between replicate samples were less than 20 percent in nearly all cases. Exceptions included one iron analysis (an RPD of 155 percent) and two manganese analyses (an RPD of 38.2 for one sample set and another set in which the one sample was 18.5 micrograms per liter (μg/L) and the replicate was reported at <0.26 μg/L) (appendix 4). In general, the replicate data indicate an acceptable level of precision.

Detections of major-ion constituents in blank samples were infrequent, at low concentrations, and indicated that field and laboratory procedures did not cause substantial bias of analytical results. Analytical results for blank samples are included in appendix 5. Major-ion (magnesium, potassium, sodium, chloride, fluoride, silica, and sulfate) and iron

Table 4. Summary of physical properties and inorganic chemistry data for groundwater samples collected by the U.S. Geological Survey in Jefferson County and surrounding areas, Montana, 2007 through 2010.

[Samples for major-ion and nutrient analysis were filtered through a 0.45-micrometer filter. Generalized geologic units (see table 2 for description): Qs; Quaternary sediment; Ts, Tertiary sediment; TKi, Tertiary through Cretaceous igneous intrusive rocks; TKv, Tertiary and Cretaceous volcanic rocks; p€u, Precambrian undifferentiated rocks. °C , degrees Celsius; --, currently (2013) no drinking-water standard for this consituent; NA, not applicable; µS/cm, microsiemens per centimeter at 25 °C; mg/L, milligrams per liter; <, less than; >, greater than; SDWR, secondary drinking water regulation; CaCO$_3$, calcium carbonate; MCL, maximum contaminant level; NO$_3$, nitrate; NO$_2$, nitrite; µg/L, micrograms per liter]

Property or constituent	Number of wells[1]	Range of values	Median value[2]	U.S. Environmental Protection Agency drinking-water regulation[3]	Number of wells and associated generalized geologic units that exceeded a drinking-water standard[4]
Physical properties					
Water temperature (°C)	128	4.5 to 44.9	10.2	--	NA
Specific conductance µS/cm at 25 °C	128	62 to 3,320	432	--	NA
Dissolved oxygen (mg/L)[5]	128	<1.0 to 17.1	5.4	--	NA
pH (standard units)	128	5.0 to 8.9	7.5	<6.5 or >8.5 (SDWR)	2 Ts, 6 TKi
Alkalinity (mg/L CaCO$_3$)	124	6.0 to 474	148	--	NA
Hardness (mg/L CaCO$_3$)	128	16.5 to 1,500	168	--	NA
Dissolved solids, total (mg/L)	128	64 to 2,000	274	500 (SDWR)	1 Qs, 8 Ts, 10 TKi, 2 p€u
Major ions, nutrients, and selected trace elements					
Calcium (mg/L)	128	2.93 to 368	46.5	--	NA
Chloride (mg/L)	128	0.39 to 975	9.02	250 (SDWR)	1 TKi
Fluoride (mg/L)	109	<0.10 to 7.76	0.36	2.0 (SDWR) / 4.0 (MCL)	2 TKi wells exceeded MCL; 1 Ts, 1 TKv, and 2 TKi wells exceeded SDWR
Magnesium (mg/L)	128	0.16 to 198	12.3	--	NA
Potassium (mg/L)	128	0.37 to 19.7	3.16	--	NA
Silica (mg/L)	128	8.23 to 64.2	22.4	--	NA
Sodium (mg/L)	128	3.34 to 323	18.2	--	NA
Sulfate (mg/L)	128	<0.18 to 1,140	51.5	250 (SDWR)	6 Ts, 2 TKi, 1 p€u
Nitrate (NO$_3$)+ Nitrite (NO$_2$) (mg/L as nitrogen)	127	<0.04 to 13.4	0.53	10 (MCL)	1 Qs, 3 TKi, 1 p€u
Iron (µg/L)	128	<6.0 to 4,450	<6.0	300 (SDWR)	1 Qs, 1 Ts, 2 TKv, 6 TKi, 2 p€u
Manganese (µg/L)	98	<0.3 to 4,190	1.75	50 (SDWR)	5 Ts, 2 TKv, 13 TKi, 2 p€u

[1]Number of wells with analytical results for a specified constituent.

[2]Based on median values of individual wells if multiple analyses exist.

[3]U.S. Environmental Protection Agency drinking-water regulation (U.S. Environmental Protection Agencey, 2009); Maximum contaminant level (MCL) is the maximum permissible level of a contaminant in water delivered to users of a public-water system. MCLs are health-based enforceable regulations. Secondary drinking water regulations (SDWRs) are non-enforceable guidelines regarding cosmetic or aesthetic effects of drinking water.

[4]Exceeded in at least one sample from an individual well.

[5]Dissolved oxygen values reported at concentrations below 1.0 mg/L are censored at <1.0 mg/L for this table.

concentrations were less than reporting levels in all eight field blank samples. Calcium was detected in one of eight field blank samples at a concentration of 0.035 milligrams per liter (mg/L), well below concentrations measured in the environmental samples which, ranged from 2.93 to 368 mg/L. Nitrate plus nitrite concentrations as well as nitrite concentrations in the four field blank samples were less than reporting levels. Barium, vanadium, and orthophosphate were analyzed in several samples collected in 2007. Only one of two field blank samples had a detected barium concentration at 0.20 µg/L; environmental sample concentrations ranged from 1.59 to 93.7 µg/L. Vanadium concentrations were less than reporting levels in the two field blank samples and orthophosphate was less than the reporting level in the one field blank sample. Manganese was analyzed in seven field blank samples; concentrations were less than reporting levels (<0.20 and <0.26 µg/L) in three of the samples and detected at concentrations ranging from 0.22 (estimated) to 0.31 µg/L in four samples (appendix 5). Manganese concentrations in the environmental samples ranged from <0.30 to 4,190 µg/L, with a median of 1.75 µg/L. As all detections for manganese in the blank samples were at or near the method reporting level, the only adjustment to the environmental concentrations was to report all estimated values below the reporting level with a remark code 'V' to indicate the value was likely affected by systematic contamination.

When major-ion concentrations were available, ionic charge balances were calculated for environmental and replicate samples. Ideally, the result of this calculation should equal zero, but in practice some deviation from zero is acceptable. An ionic charge balance within plus or minus 5 percent is generally considered acceptable (Clesceri and others, 1998). If significant deviation from zero occurs, there must be either errors in the analytical measurement or the presence of an ionic specie or species at substantial concentrations that were not included in the analysis. The range of values for the cation/anion balance calculation for the 166 samples with nearly complete major-ion data collected in this study was 6.9 percent below to 9.34 percent above balance, with only 7 samples greater than 5 percent (ranging from 5.2 to 9.3 percent) and only 2 less than -5 percent (-5.3 and -6.9 percent). Nearly 95 percent (157 of 166 samples) of ionic charge balances were within plus or minus 5 percent. Given the range of values for the samples analyzed in this study, we consider all samples to have an acceptable cation/anion balance; therefore, any analytical errors present are small enough not to affect the interpretation of the data, and all major dissolved ionic species were included in the analyses.

Occurrence and Distribution of Radiochemical Constituents in Groundwater

Radiochemical constituents including gross alpha-particle activity and gross beta-particle activity, and concentrations of uranium, radium, radon, and polonium from the 128 wells sampled for this study are tabulated in appendix 6 and summarized in table 5. Concentrations of radium-226 were analyzed by two techniques, and results of both techniques are included in appendix 6. Radioactive constituents were detected in each of the wells sampled for this study (168 total samples) regardless of location or geologic unit. Overall, nearly 41 percent (52 of the 128) of the sampled wells had at least one constituent with a concentration that exceeded a MCL for uranium or combined radium-226 and radium-228, a proposed MCL for radon (4,000 pCi/L), or screening levels for gross alpha-particle and gross beta-particle activity.

Radioactive constituents exceeded at least one USEPA MCL, proposed MCL, or screening level in samples from at least one well within five of six generalized geologic units assessed during this study (table 5). Established or proposed drinking-water standards were exceeded most frequently in water from wells completed in the generalized geologic unit (TKi) that includes rocks of the Boulder batholith and other Tertiary through Cretaceous igneous intrusive rocks (commonly described as granite). At least one drinking-water standard or proposed drinking-water standard for radioactive constituents was exceeded in water from 38 of 62 TKi sampled wells. The 30 µg/L MCL for uranium was exceeded in 15 (24 percent) of 62 sampled TKi wells. The 4,000 pCi/L proposed alternative MCL for radon was exceeded in 31 (50 percent) of 62 sampled TKi wells. The 5 µg/L MCL for combined radium-226 and radium-228 was exceeded in 10 (27 percent) of 37 TKi wells sampled for those constituents. Only samples from the generalized Cretaceous through Cambrian undifferentiated sedimentary rocks unit (KЄu) had measured concentrations and activities that were less than associated water-quality criteria. However, only three samples were collected from the KЄu and this unit may warrant further evaluation.

Table 5.   Summary of radiochemistry data for groundwater samples collected by the U.S. Geological Survey in Jefferson County and surrounding areas, Montana, 2007 through 2010.

[Samples for uranium, radium-224, radium-226, radium-228, polonium-210, gross alpha-particle radioactivity, and gross beta-particle radioactivity analysis were filtered through a 0.45-micrometer filter. Radon samples were unfiltered. Generalized geologic units (see table 2 for description): Qs; Quaternary sediment; Ts, Tertiary sediment; TKi, Tertiary through Cretaceous igneous intrusive rocks; TKv, Tertiary and Cretaceous volcanic rocks; KЄu, Cretaceous through Cambrian undifferentiated rocks; pЄu, Precambrian undifferentiated rocks. LRL, laboratory reporting level; ssLc, sample specific critical level; µg/L, micrograms per liter; <, less than reporting level; USEPA, U.S. Environmental Protection Agency; MCL, maximum contaminant level; pCi/L, picocuries per liter; mrem/yr, millirem per year; CA-MCL, California maximum contaminant level; --, currently (2013) no drinking-water standard for this individual consituent; NA, not applicable]

Analyte	Number of wells[1]	Range of LRLs or ssLcs	Range of values[2]	Median value[3]	Drinking-water standard or screening level[4]	Number of wells exceeding drinking-water standard or screening level[5]	Comments
Uranium, natural (µg/L)	128	0.01 to .04	<0.04 to 1,130	4.37	30 ug/L (USEPA MCL)	18	At or above MCL in 1 of 18 Qs wells, 2 of 24 Ts wells, 15 of 62 TKi wells, 0 of 12 TKv wells, 0 of 3 KЄu, and 0 of 9 pЄu wells.
Radon-222 (pCi/L)	127	10 to 36	50 to 45,000	1,940	300 pCi/L (proposed USEPA MCL), 4,000 pCi/L (proposed alternative USEPA MCL)[6]	34	At or above the proposed alternative MCL in 1 of 18 Qs wells, 2 of 24 Ts wells, 31 of 62 TKi wells, 0 of 11 TKv wells, 0 of 3 KЄu, and 0 of 9 pЄu wells. Samples from 121 wells exceeded the lower proposed MCL of 300 pCi/L.
Gross alpha-particle activity, 72-hour count (pCi/L)	104	0.24 to 4.4	<ssLc to 740	9.20	15 pCi/L (screening level)[7]	42	At or above screening level in 2 of 13 Qs wells, 3 of 16 Ts wells, 33 of 56 TKi wells, 1 of 9 TKv wells, 0 of 1 KЄu wells, and 3 of 9 pЄu wells.
Gross alpha-particle activity, 30-day count (pCi/L)	104	0.22 to 3.7	<ssLc to 880	5.83	15 pCi/L (screening level)[7]	34	At or above screening level in 1 of 13 Qs wells, 3 of 16 Ts wells, 27 of 56 TKi wells, 1 of 9 TKv wells, 0 of 9 KЄu wells, and 2 of 9 pЄu wells.
Gross beta-particle activity, 72-hour (pCi/L)	104	0.40 to 1.7	<ssLc to 57	4.75	4 mrem/yr (USEPA MCL); 50 pCi/L (CA-MCL)[8]	2	At or above screening level in 0 of 13 Qs wells, 0 of 16 Ts wells, 2 of 56 TKi wells, 0 of 9 TKv wells, 0 of 1 KЄu wells, and 0 of 9 pЄu wells.
Gross beta-particle activity, 30-day count (pCi/L)	104	0.42 to 1.4	<ssLc to 323	6.70	4 mrem/yr (USEPA MCL); 50 pCi/L (CA-MCL)[8]	5	At or above screening level in 0 of 13 Qs wells, 0 of 16 Ts wells, 5 of 56 TKi wells, 0 of 9 TKv wells, 0 of 1 KЄu wells, and 0 of 9 pЄu wells.
Radium-224 (pCi/L)	39	0.064 to 1.0	<ssLc to 16.1	0.80	--	--	--
Radium-226 (pCi/L), alpha spectrometry method	40	0.031 to 0.22	<ssLc to 24.0	0.95	--	--	--
Radium-226 (pCi/L), radon emanation method	46	0.011 to 0.10	<ssLc to 22.1	0.51	--	--	--
Radium-228 (pCi/L)	47	0.17 to 0.54	<ssLc to 19.0	0.73	--	--	--
Combined radium-226 and radium-228 (pCi/L)[9,10]	47	NA[9]	<ssLc to 39.1	1.40	5.0 (USEPA MCL)	10	Above MCL in 10 of 37 sampled TKi wells.

Table 5. Summary of radiochemistry data for groundwater samples collected by the U.S. Geological Survey in Jefferson County and surrounding areas, Montana, 2007 through 2010.—Continued

[Samples for uranium, radium-224, radium-226, radium-228, polonium-210, gross alpha-particle radioactivity, and gross beta-particle radioactivity analysis were filtered through a 0.45-micrometer filter. Radon samples were unfiltered. Generalized geologic units (see table 2 for description): Qs; Quaternary sediment; Ts, Tertiary sediment; TKi, Tertiary through Cretaceous igneous intrusive rocks; TKv, Tertiary and Cretaceous volcanic rocks; KЄu, Cretaceous through Cambrian undifferentiated rocks; pЄu, Precambrian undifferentiated rocks. LRL, laboratory reporting level; ssLc, sample specific critical level; µg/L, micrograms per liter; <, less than reporting level; USEPA, U.S. Environmental Protection Agency; MCL, maximum contaminant level; pCi/L, picocuries per liter; mrem/yr, millirem per year; CA-MCL, California maximum contaminant level; --, currently (2013) no drinking-water standard for this individual consituent; NA, not applicable]

Analyte	Number of wells[1]	Range of LRLs or ssLcs	Range of values[2]	Median value[3]	Drinking-water standard or screening level[4]	Number of wells exceeding drinking-water standard or screening level[5]	Comments
Polonium-210 (pCi/L)	40	0.013 to 0.072	<ssLc to 3.08	0.032	--	--	Concentrations greater than the ssLc were reported in 27 of 40 wells, 26 of the samples considered as detections were collected from wells completed in the TKi. One of 4 Ts wells and 2 of 31 TKi wells were above 1 pCi/L.

[1]Number of wells with analysis of specified constituent.

[2]Range of values for all analyses.

[3]Median value of all sampled wells; the median values of wells with multiple samples were used in this calculation.

[4]Maximum contaminant level (MCL) is the maximum permissible level of a contaminant in water delivered to users of a public-water system. MCLs are health-based enforceable regulations (U.S. Environmental Protection Agency, 2000). Screening level—a reference level used for compliance monitoring and subsequent sampling requirements (U.S. Environmental Protection Agency, 2000); CA-MCL, California Department of Public Health drinking water standard (California Department of Public Health, 2008).

[5]Exceeded in at least one sample from a well.

[6]Proposed U.S. Environmental Protection Agency MCL for states that choose to develop enhanced state programs to address the health risks from radon in indoor air—known as Multimedia Mitigation (MMM) programs. If a state chooses not to develop an MMM program, individual water systems in that state would be required to either reduce radon in their system's drinking water to the proposed MCL of 300 pCi/L or develop individual local MMM programs and reduce levels in drinking water to the alternaive MCL of 4,000 pCi/L.

[7]U.S. Environmental Protection Agency (2000) requires that community water systems that have gross alpha-particle activity levels greater than 15 pCi/L will be required to monitor for uranium.

[8]U.S. Environmental Protection Agency (2000) gross beta-particle activity MCL of 4 mmrem/yr as it applies to annual dose equivalent to the total body or any internal organ from man-made beta particle and photon emitters. The California Department of Public Health drinking water standard (CA-MCL) for gross beta-particle activity is listed as 4 mmrem/yr and 50 pCi/L (California Department of Public Health, 2008). A gross beta-particle activity of 50 pCi/L is set as a screening level for this report.

[9]Radium-226 or radium-228 values reported as less than ssLc were set equal to 0 for this calculation.

[10]Radium-226 analyzed by the alpha spectrometry method was substituted for this calculation for one site (well 463344112104701) for which the radon emanation method result was not available.

Quality Assurance Results of Radionuclides

Twelve sets of replicate samples were analyzed for radio-chemical constituents (appendix 7). Concentration data for the replicate samples generally indicated good agreement for most of the radiochemical analytes. The largest RPDs were typically for replicate samples with concentrations near reporting levels or associated ssLcs. The RPDs were less than 20 percent or the difference in concentrations were less than 1 pCi/L or both for each replicate for all analytes except gross alpha-particle activity (72-hour count) (appendix 7). The RPDs for gross alpha-particle activity (72-hour count) exceeded 20 percent in 2 of 9 replicates (19 and 29 pCi/L with an RPD of 20.8 percent; 16 and 30 pCi/L with an RPD of 30.4 percent) indicating more variability in the analytical methods than anticipated. However, the results were not consistent between replicates indicating there may be other sources of variability rather than the laboratory analysis.

Detections of radiochemical constituents in blank samples were infrequent, at low concentrations, and indicated that field and laboratory procedures did not cause substantial bias of analytical results. Results of radiochemical analyses of blank samples are included in appendix 8. Dissolved uranium concentrations were less than reporting levels (0.04 and 0.008 µg/L) in all seven field blank samples. Radon results were considered nondetects in the two field blanks. All gross alpha-particle activity values (72-hour and 30-day counts; 5 samples) were either considered nondetects or had activities of 0.43 pCi/L or less, which are values much less than the 9.20 pCi/L and 5.83 pCi/L median values (72-hour and 30-day counts, respectively) observed in the environmental samples. Gross beta-particle activity (72-hour and 30-day counts), and polonium-210, radium-224, radium-226 (radon emanation method), radium-228, uranium-234, and uranium-235 concentrations were all measured at levels considered as nondetects. Radium-226 (alpha spectrometry method) had a reported concentration of 0.2 pCi/L and uranium-238 had a reported concentration of 0.015 pCi/L from the field-blank sample analyzed for those constituents. Median environmental concentrations for radium-226 (alpha spectrometry method) and uranium-238 were 0.95 pCi/L and 5.4 pCi/L, respectively.

Uranium

Uranium was detected in 125 of the 128 wells sampled for this study (appendix 6). Concentrations ranged from less than 0.04 µg/L to 1,130 µg/L, with a median of 4.37 µg/L (table 5). Uranium concentrations in at least 1 sample from 18 of 128 wells (14 percent of the sampled wells) exceeded the USEPA MCL of 30 µg/L. The geographic distribution of uranium concentrations in groundwater from the 128 wells sampled for this study and 441 wells in Jefferson County and surrounding counties included in the MBMG GWIC database is presented in figure 5; median values are represented for wells sampled more than once. The range and median uranium

concentrations of samples collected for this study from each of the six generalized geologic units are shown in figure 6.

Uranium was detected in groundwater from all generalized geologic units sampled for this study. Concentrations greater than the MCL were observed in samples from Qs (1 well), Ts (2 wells), and TKi (15 wells) (fig. 6). Eight of the 10 highest uranium concentrations were from study wells completed in TKi. Overall, study wells from the Boulder batholith and similar geologic units (TKi) had the highest median concentration (8.49 µg/L). Within the GWIC database, the 10 wells with the highest median uranium concentrations (the highest median concentrations of wells sampled more than once) were completed in Qs (2 wells), Ts (5 wells), TKi (2 wells), and p€u (1 well). The highest uranium value (144.7 ug/L) of the data from domestic wells retrieved from the GWIC database (accessed on February 7, 2012, at *http://mbmggwic.mtech.edu/*) was from a well completed in Ts located in Deer Lodge County.

Uranium concentrations from this study (median value of 4.37 µg/L) appear elevated in comparison to national values. Ayotte and others (2011) determined that the median concentration of uranium in 3,541 NAWQA groundwater samples collected between 1992 and 2003 was 0.52 µg/L. A more extensive dataset of 4,286 water supply wells sampled between 1992 and 2012 within 48 states for the NAWQA program had uranium concentrations ranging from less than 1.0 µg/L to 1,415 µg/L, with a median of less than 1.0 µg/L (U.S. Geological Survey, 2012b). For the two NAWQA datasets, only about 3–4 percent of the wells exceeded the MCL for uranium, whereas about 18 percent of the wells sampled for this study exceeded the MCL.

Median uranium concentrations in samples from 62 wells from this study as well as sample results from 64 wells retrieved from the GWIC database (accessed on February 7, 2012, at *http://mbmggwic.mtech.edu*) that were all completed in the TKi ranged from less than 1 µg/L to 890.5 µg/L, with an overall median value of 10.5 µg/L. The spatial distribution of these wells is not even throughout the study area, and a visual inspection of the distribution of uranium concentrations (fig. 5) gives the appearance that uranium concentrations may be elevated in certain areas or well clusters. This possibility of high-concentration areas may be a function of the increased density (number of wells) sampled in these areas and the inclusion of some wells that were targeted rather than randomly selected. For example, wells within a 5-mile radius of the town of Clancy in northern Jefferson County appear to be elevated. Of the analyses of 126 wells completed in TKi from this study and from the GWIC database, samples within 5.0 miles of Clancy had four measured concentrations that were among the top eight within the overall area. However, the median uranium value of 11.6 µg/L (with an associated interquartile range of 7.2 to 43.7 µg/L) for the 19 wells within a 5-mile radius of Clancy was only slightly higher than the median value of 9.86 µg/L (with an associated interquartile range of 3.63 to 17.9 µg/L) for the 107 wells outside of the Clancy area. Results of the rank-sum statistical test indicated that most uranium concentrations

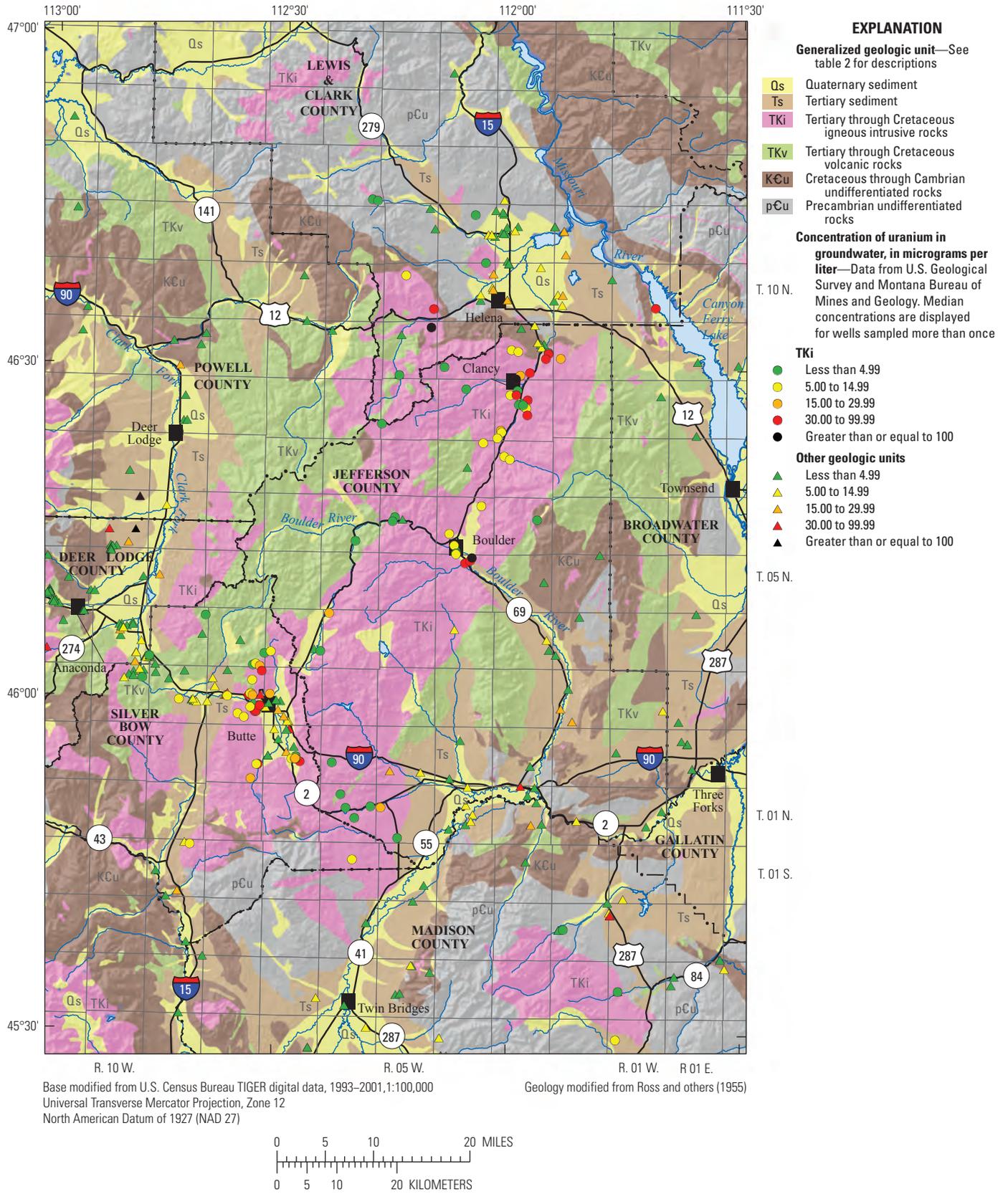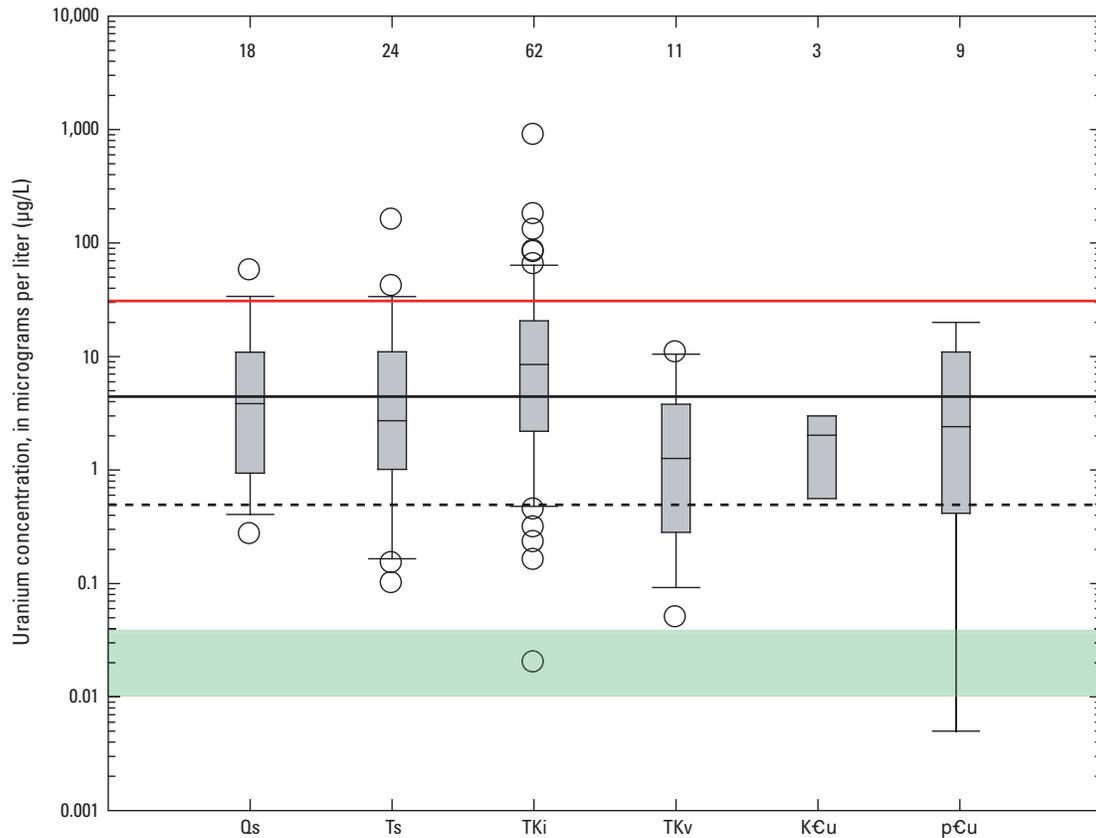**Figure 5.**   Magnitude and distribution of uranium concentrations in groundwater samples from Jefferson County and surrounding areas, Montana, 1976 through 2011.

Figure 6.   Uranium concentrations in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. Data collected by the U.S. Geological Survey. Median values are displayed for wells sampled more than once.

for sampled wells within 5 miles of Clancy were not significantly different (*p*-value of 0.29) from samples collected from TKi wells in the remainder of the study area.

Radon

Radon was detected in each of the 127 wells sampled for radon analysis for this study (table 5, appendix 6). Concentrations ranged from 50 to 45,000 pCi/L, with a median of 1,940 pCi/L. Concentrations in samples from 121 (95 percent) of the study wells from all of the generalized geologic units exceeded the proposed 300 pCi/L USEPA MCL. Radon concentrations in at least one sample from 34 wells (27 percent of the sampled wells) exceeded the proposed USEPA Alternative Maximum Contaminant Level (AMCL) of 4,000 pCi/L. The geographic distribution of radon concentrations is presented in figure 7 and includes data from 227 wells retrieved from the GWIC database (accessed on February 7, 2012, at *http://mbmggwic.mtech.edu*) with concentrations ranging from less than 10 to 54,100 pCi/L.

Radon concentrations greater than the proposed 4,000 pCi/L AMCL were observed in samples from Qs (1 well), Ts (2 wells), and TKi (31 wells) (table 5, fig. 8). Nine of the 10 highest radon concentrations were from wells completed in TKi; a single well completed in Qs also was among the 10 highest (fig. 8). Overall study wells from the Boulder batholith and similar geologic units (TKi) had the highest median radon concentration (3,860 pCi/L) (fig. 8). The 10 wells with the highest radon concentrations (the highest median concentrations of wells sampled more than once) within the GWIC database include wells completed in Ts (2 wells), TKi (7 wells), and in TKv (1 well). The highest radon (54,100 pCi/L) value within the GWIC database was a sample from a well in Powell County completed in Ts.

Radon concentrations from this study (median value of 1,940 pCi/L) appear elevated in comparison to national values. Ayotte and others (2011) determined that concentrations in 2.7 percent of 3,877 samples from NAWQA wells sampled between 1992 and 2003 exceeded the proposed AMCL of 4,000 pCi/L for radon with an overall median concentration of 430 pCi/L. An extensive dataset of 3,622 wells sampled between 1991 and 2011 within 48 states for the NAWQA program (U.S. Geological Survey, 2012b) had radon concentrations ranging from less than 80 pCi/L to 101,760 pCi/L, with a median of 403 pCi/L (99.5 percent of the samples were less than 10,000 pCi/L). Approximately 2.7 percent (100 of the 3,622) of the NAWQA wells had at least one analytical result that exceeded the proposed 4,000 pCi/L AMCL for radon, whereas about 27 percent of the wells for this study had sample concentrations that exceeded the AMCL.

Sample results from 62 wells collected as part of this study and results from 49 wells retrieved from the GWIC database (accessed on February 7, 2012, at *http://mbmggwic.mtech.edu*) that were all completed in the TKi had radon concentrations ranging from less than 80 pCi/L to 43,900 pCi/L, with a median value of 3,010 pCi/L. The spatial distribution of these wells is not even throughout the study area, and a visual inspection of the distribution of radon concentrations (fig. 7) suggests radon concentrations are elevated in certain areas or well clusters. For example, radon concentrations in water from wells within a 5-mile radius of the town of Clancy in northern Jefferson County appear to be elevated. The median radon value of 9,648 pCi/L (with an associated interquartile range of 5,990 to 11,400 pCi/L) for the 17 wells within a 5-mile radius of Clancy was higher than the median value of 2,290 pCi/L (with an associated interquartile range of 850to 6,028 pCi/L) for the 94 wells outside of the Clancy area. Results of the rank-sum statistical test indicated that most radon concentrations from sampled wells within 5 miles of Clancy were significantly different (*p*-value of <0.05) from samples collected from TKi wells in the remainder of the study area.

Gross Alpha-Particle Activity

Gross alpha-particle activity is a measure of the total amount of radioactivity in a water sample attributable to the radioactive decay of alpha-emitting elements. Laboratory results for gross alpha-particle activities from this study are reported as actual gross alpha-particle activity (table 5, appendixes 6, 7, and 8). In terms of compliance monitoring for public water-supply systems, U.S. Environmental Protection Agency (2000) requires uranium monitoring for community water systems that have gross alpha-particle activity levels greater than 15 pCi/L. The USEPA Maximum Contaminant Level is listed as 15 picocuries per liter (pCi/L) for adjusted gross alpha-particle activity, which is defined as the gross alpha-particle activity minus activity caused by uranium and radon (U.S. Environmental Protection Agency, 2000). Gross alpha-particle activities reported from the laboratory were not adjusted (reduced) for activity contributions by uranium (activity caused by radon was effectively removed during the analytical process), therefore they are conservatively high estimates relative to the regulated adjusted gross alpha-particle activity. Gross alpha-particle activity was greater than the respective ssLcs in samples from 102 of 104 wells (72-hour count) and 92 of 104 wells (30-day count) analyzed for this study (appendix 6).

The gross alpha-particle activities (72-hour count) ranged from less than the ssLcs in 2 wells to 740 pCi/L, with a median of 9.20 pCi/L; activities in samples from 42 wells exceeded the 15 pCi/L screening level (table 5). The gross alpha-particle activity (30-day count) ranged from less than the ssLcs in 12 wells to 880 pCi/L, with a median of 5.83 pCi/L; activities in samples from 34 wells exceeded 15 pCi/L (table 5, appendix 6). As study data indicate (figs. 9 and 10), the holding time between sample collection and analysis can affect measured activities, with a general decrease in gross alpha-particle activities with time. Typically, gross alpha-particle activity decreased about 37 percent between measurements at 72 hours and 30 days (fig. 10). This decrease in gross alpha-particle activity between the 72-hour and 30-day counts indicates the presence of short-lived (decay quickly) alpha-emitting radionuclides in the sampled waters.
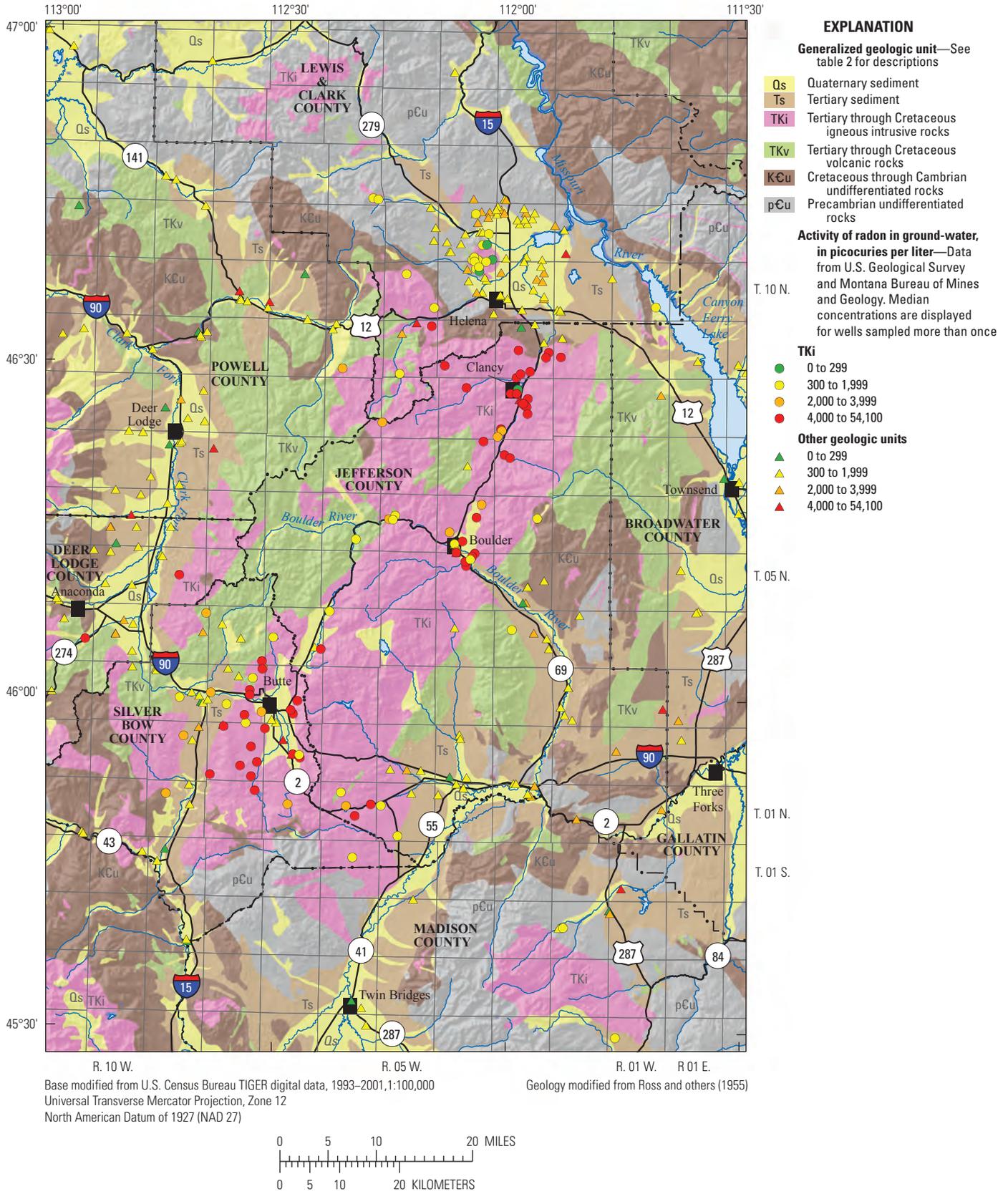
Figure 7. Magnitude and distribution of radon concentrations in groundwater samples from Jefferson County and surrounding areas, Montana, 1992–2011.
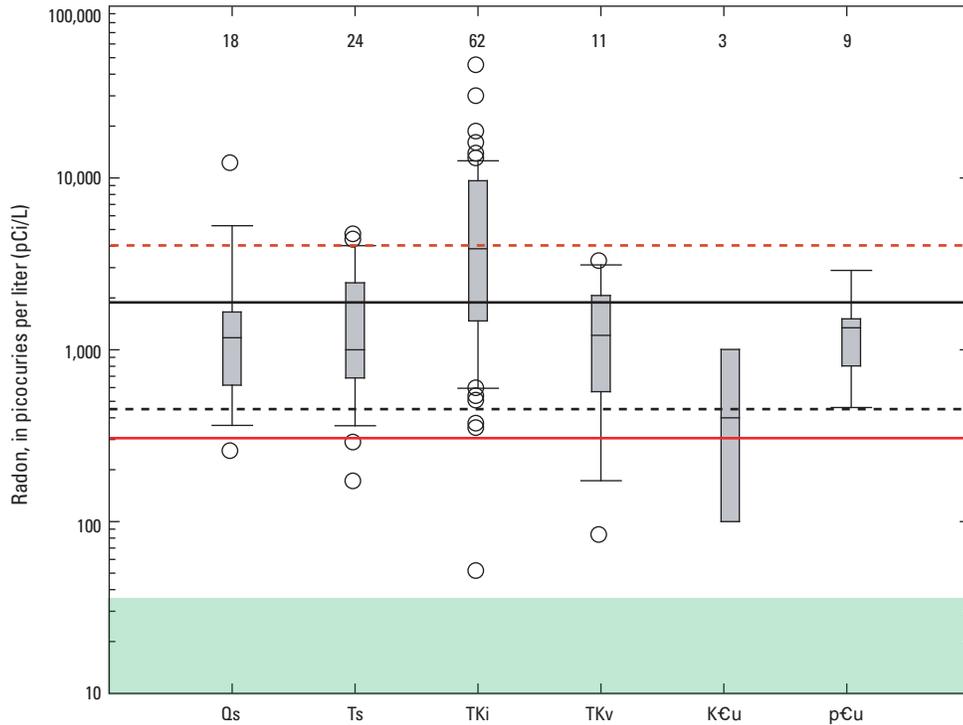
EXPLANATION

Generalized geologic unit—See table 2 for descriptions

Qs—Quaternary sediment

Ts—Tertiary sediment

TKi—Tertiary through Cretaceous igneous intrusive rocks

TKv—Tertiary through Cretaceous volcanic rocks

KЄu—Cretaceous through Cambrian undifferentiated rocks

pЄu—Precambrian undifferentiated rocks

18   **Number of samples**

○   **Outlier**

    90th percentile

    75th percentile

    Median (50th percentile)

    25th percentile

    10th percentile

○   **Outlier**

    Minimium reporting limits of samples collected for this study (10 to 36 pCi/L)

    Proposed maximum contaminant level (300 pCi/L) (U.S. Environmental Protection Agency, 1999c) and State of Montana water quality standard (Montana Department of Environmental Quality, 2012)

- - -   **Proposed alternative maximum contaminant level (4,000 pCi/L) (U.S. Environmental Protection Agency, 1999c)**

    Median concentration of all samples collected for this study (1,940 pCi/L)—Median values of wells with multiple samples were used in this calculation

- - -   **Median concentration of national data set from the U.S. Geological Survey National Water Quality Assessment Program (430 pCi/L) (Ayotte and others, 2011)**

Figure 8.   Radon concentrations in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. Data collected by the U.S. Geological Survey. Median values are displayed for wells sampled more than once.

Figure 9.   Gross alpha-particle activity (72-hour and 30-day counts) in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. Median values are displayed for wells sampled more than once. Values are displayed as reported from the laboratory and have not been adjusted to remove uranium activity.
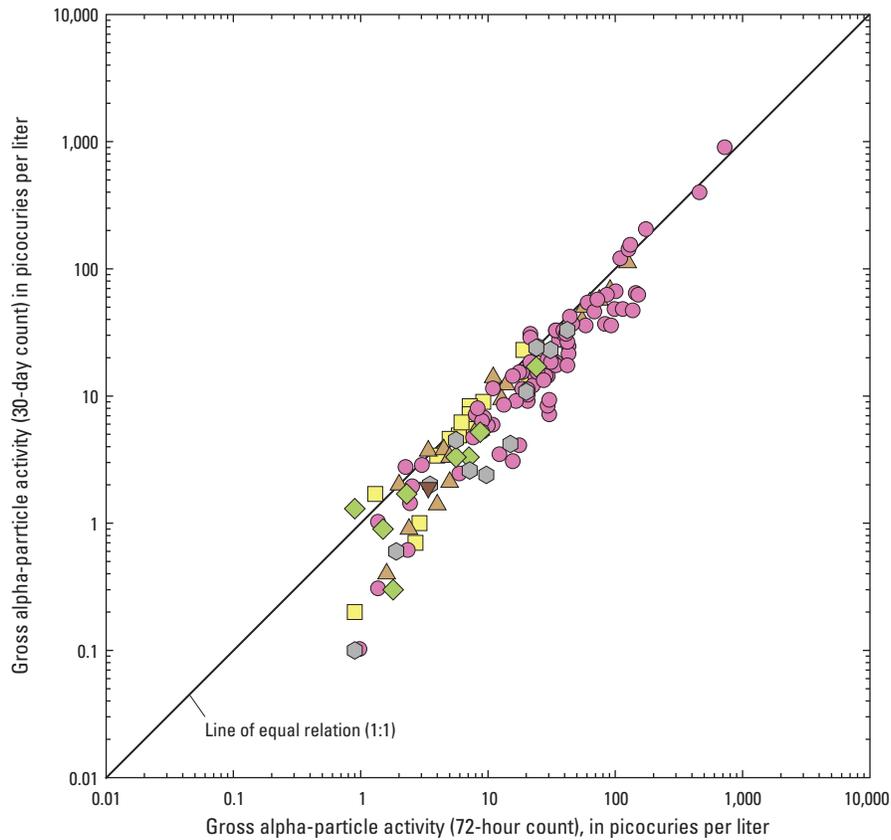
Figure 10.   Relation between gross alpha-particle activity at 72-hour and 30-day counts in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. Values are displayed as reported from the laboratory and have not been adjusted to remove uranium activity.

The adjusted gross alpha-particle activity (calculated activity after subtracting activity associated with uranium based on a 0.67 picocurie per microgram conversion factor) exceeded the USEPA MCL of 15 pCi/L in samples from 26 wells at the 72-hour count and 13 wells at the 30-day count. Uranium concentrations were determined for all samples analyzed for this study and were reported in micrograms per liter. To convert uranium concentration in micrograms per liter to activity in picocuries per liter for compliance evaluations, the uranium concentration in micrograms per liter can be multiplied by a factor of 0.67 (U.S. Environmental Protection Agency, 2000) based on the assumption that uranium-238 and uranium-234 are present at a 1:1 ratio. Adjusted gross alpha-particle activities were calculated in this manner, but it is understood that this conversion may over- or under-estimate actual uranium activities. Uranium isotopes (U-234, U-235, and U-238) reported as activities in picocuries per liter also were analyzed from 52 samples. Study data indicate that samples with uranium isotope activities (reported as picocuries per liter) and dissolved uranium concentrations (reported as micrograms per liter) had concentration to activity ratios ranging from 0.5 to 2.2, with a mean value of 0.8 (slightly higher than the 0.67 factor used for uranium concentration to activity conversions acceptable by USEPA). Therefore, the 0.67 conversion factor is considered a conservative approximation of the uranium activity for this study dataset.

Gross alpha-particle activity was detected in groundwater from all geologic units sampled for this study (fig. 9). Samples from wells completed in the Boulder batholith or similar rocks (TKi) had the highest median gross alpha-particle activities [22.5 pCi/L (72-hour count) and 11.3 pCi/L (30-day count); calculated using median values of individual wells sampled more than once]. Median gross alpha-particle activities (72-hour count) of samples from Qs, Ts, TKv, and pЄu were lower (6.2, 6.5, 2.3, and 7.2 pCi/L, respectively). About 40 percent (42 of 104 wells) of the wells had gross alpha-particle activities (72-hour count) at or greater than the screening level of 15 pCi/L, and about 30 percent (31 of 104 wells) had gross alpha-particle activities (30-day count) at or greater than this screening level. Gross alpha-particle activities at or greater than the screening level were most common in samples from wells completed in the TKi but also were measured in samples from wells completed in Qs, Ts, TKv, and pЄu (table 5). Only one well within the KЄu was analyzed for gross alpha-particle activity (3.4 pCi/L, 72-hour count).

Gross alpha-particle activities from this study appear elevated in comparison to national values. A set of 248 wells sampled between 2004 and 2011 within 15 states for the NAWQA program had gross alpha-particle activities (72-hour count) ranging from less than the ssLc to 654 pCi/L, with a median of 2.33 pCi/L (U.S. Geological Survey, 2012b). Activities in approximately 15 percent of these wells exceeded the 15 pCi/L screening level for gross alpha-particle activity (72-hour count), whereas activities in about 40 percent of the wells sampled for this study exceeded this screening level.

Gross Beta-Particle Activity

Gross beta-particle activity is a measure of the total amount of radioactivity in a water sample attributable to the radioactive decay of beta-emitting elements. The gross beta-particle activity (72-hour count) was greater than the associated ssLc in 99 of 104 samples, with activities as high as 57 pCi/L and a median of 4.75 pCi/L (table 5, fig. 11, appendix 6). Gross beta-particle activity (30-day count) was higher than the ssLc in all but 1 of the 104 wells, with activities that ranged from the ssLc to 323 pCi/L and a median of 6.70 pCi/L (table 5). Two of 104 wells had gross beta-particle activities (72-hour count) at or greater than the screening level of 50 pCi/L, and 5 of the 104 wells had at least 1 sample at or greater than the screening level (30–day count).

Gross beta-particle activity was detected in groundwater from all geologic units sampled for this study (fig. 11). Only samples from wells that were completed in the Boulder batholith or similar rocks (TKi) had gross beta-particle activities at or greater than the 50 pCi/L screening level. Median gross beta-particle activities from the wells completed in the TKi and Ts were the highest [TKi, 5.63 pCi/L (72-hour count) and 8.08 pCi/L (30-day count); Ts, 5.65 pCi/L (72-hour count) and 7.50 pCi/L (30-day count)].

Gross beta-particle activities from this study (median values of 4.75 pCi/L and 6.7 pCi/L, 72-hour and 30-day count, respectively) appear elevated in comparison to national values. A set of 248 wells sampled between 2004 and 2011 within 15 states for the NAWQA program had gross beta-particle activities ranging from less than the ssLc to 37.6 pCi/L, with a median of 2.5 pCi/L (72-hour count), and less than the ssLc to 461.0 pCi/L, with a median of 2.9 pCi/L (30-day count) (U.S. Geological Survey, 2012b). Less than 1 percent of the NAWQA wells exceeded the 50 pCi/L screening level for gross beta-particle activity (30-day count), whereas about 5 percent of the wells sampled for this study exceeded this screening level.

The holding time between sample collection and analysis can affect measured activities. Gross beta-particle activity increased on average about 31 percent in samples measured at 72 hours and again at 30 days (activities in 110 samples increased, 22 decreased, and 6 stayed about the same between the 72-hour and 30-day counts) (fig. 12). This increase in gross beta-particle activity over that time period usually indicates the ingrowth (incremental accumulation of daughter products generated by the decay of the parent) of beta-emitting progeny of uranium. Welch and others (1995) examined the analyses of about 800 groundwater samples within the United States and determined that the ingrowth of beta-emitting progeny of uranium-238 (specifically thorium-234 and palladium-234, which are relatively short-lived beta-particle emitters) contributed much of the gross beta-particle activity measured in their samples.

Figure 11.   Gross beta-particle activity (72-hour and 30-day count) in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. Median values are displayed for wells sampled more than once.

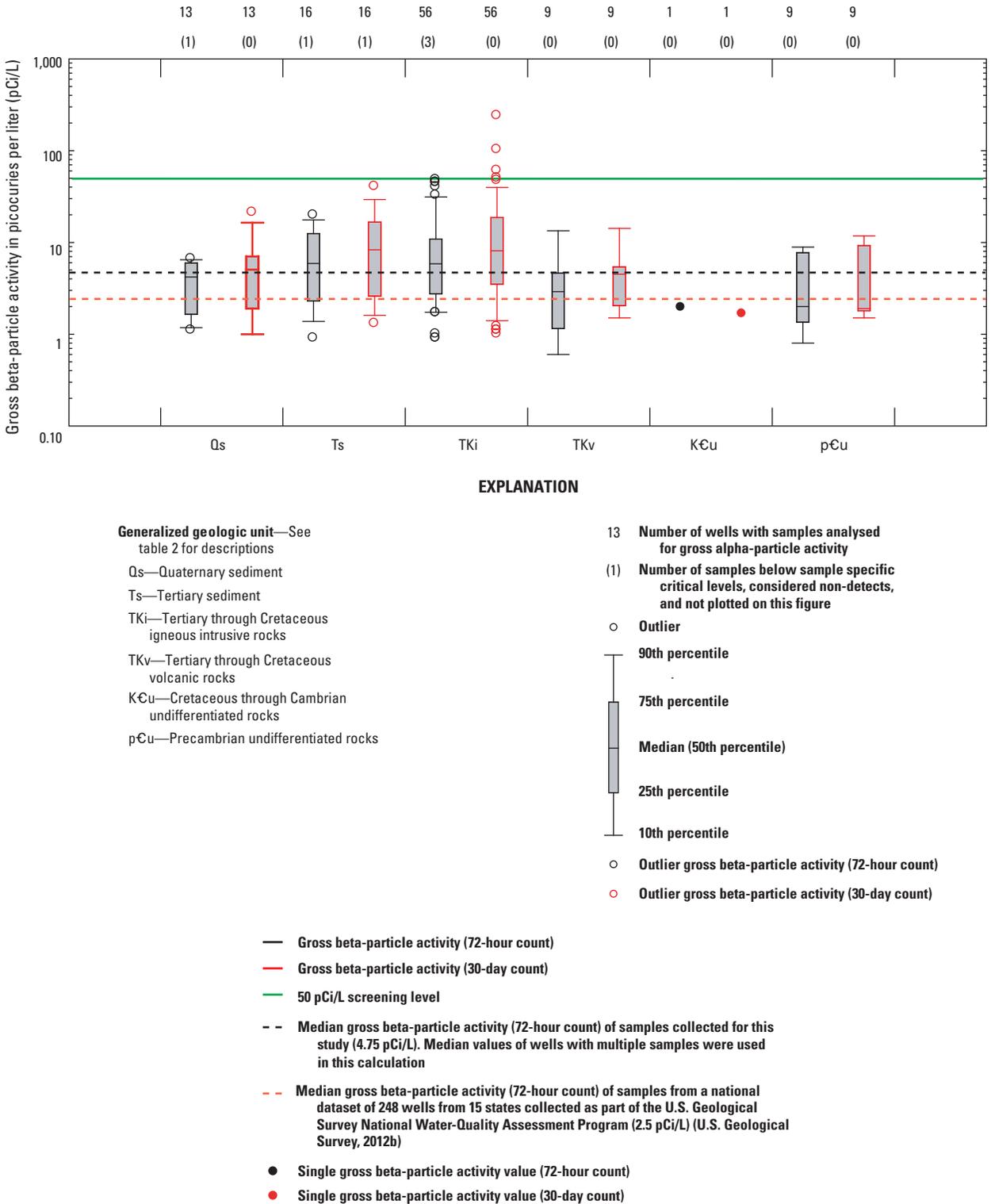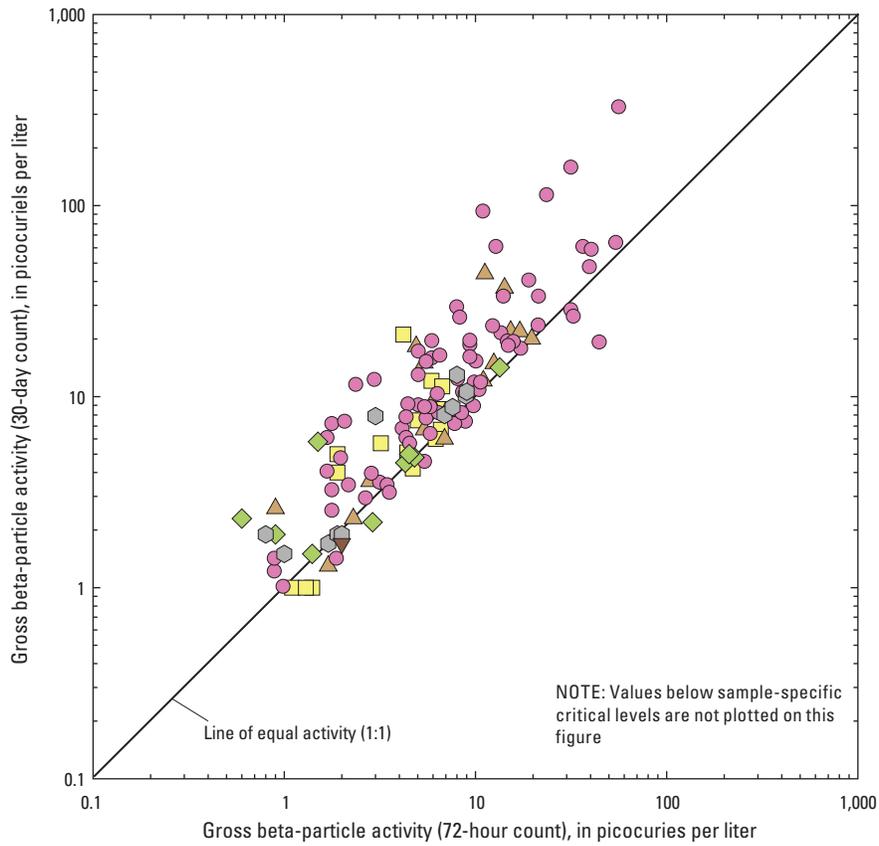**Figure 12.** Relation between gross beta-particle activity at 72-hour and 30-day counts in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Radium

Radium isotopes (radium-224, radium-226, and radium-228) were analyzed in samples from a subset of wells where either elevated radioactive constituents had been measured or had been measured in the vicinity. The radium MCL is defined as 5 pCi/L for the sum of the radium isotopes, radium-226 and radium-228. Of the 47 wells that had samples with both radium-226 and radium-228 analyzed as part of this study, 10 of the wells exceeded the combined radium-226 and radium-228 MCL of 5 pCi/L.

Radium-224 was detected in 32 of the 39 wells and concentrations ranged from less than the ssLc to 16.1 pCi/L, with a median of 0.80 pCi/L (table 5, appendix 6). Radium-224 concentrations from this study appear elevated in comparison to a set of 486 wells sampled between 1998 and 2011 within 25 states for the NAWQA program. The NAWQA samples had radium-224 values ranging from less than the ssLc to 13.9 pCi/L, with a median of 0.1 pCi/L (U.S. Geological Survey, 2012b). Although there is no current (2013) MCL for radium-224, Parsa (1998) calculated that radium-224 could contribute considerable gross alpha-particle activity to a water sample for about 7 days after sample collection.

Radium-226 concentrations in some samples were analyzed using two methods (alpha spectrometry and radon emanation; table 3). Results from the two methods were comparable, with similar ranges of values and similar median values (table 5). Relative percent differences were less than 31 percent between the results of the two methods from individual wells with radium-226 concentrations greater than 0.5 pCi/L. The radon emanation method is an accepted USEPA method when comparing the sum of radium-226 and radium-228 to the USEPA MCL (U.S. Environmental Protection Agency, 2008). Therefore, the following discussion refers to radium-226 values analyzed using the radon emanation method. Radium-226 was detected in 44 of the 46 wells with concentrations that ranged from less than the ssLc to 22.1 pCi/L and a median concentration of 0.51 pCi/L (table 5). Radium-226 values from this study appear elevated in comparison to a set of 1,480 samples from 1,425 wells sampled between 1992 and 2011 within 44 states as part of the NAWQA program. The NAWQA samples had radium-226 values ranging from less than the ssLc to 52.8 pCi/L, with a median of 0.14 pCi/L (U.S. Geological Survey, 2012b). Only one sample from the NAWQA dataset had a measured radium-226 concentration (52.8 pCi/L) higher than the highest value measured in this study (22.1 pCi/L).

Radium-228 was detected in 39 of the 47 wells and concentrations ranged from less than the ssLc to 19.0 pCi/L, with a median of 0.73 pCi/L (table 5). Radium-228 values from six of the wells exceeded the 5 pCi/L MCL for combined radium. Radium-228 results from this study appear slightly elevated in comparison to a set of 1,492 samples from 1,439 wells sampled between 1992 and 2011 within 43 states for the NAWQA program. The NAWQA samples had radium-228 concentrations ranging from less than the ssLc to 16.2 pCi/L, with a median of 0.46 pCi/L (U.S. Geological Survey, 2012b).

The combined radium-226 and radium-228 concentrations for the 47 wells sampled for this study ranged from less than the ssLcs to 39.1 pCi/L, with a median of 1.40 pCi/L. Ten of the wells exceeded the combined radium-226 and radium-228 MCL of 5 pCi/L. Although most wells sampled for radium (36 of 47 wells) were completed in the TKi geologic unit, individual samples from the Ts and pЄu geologic units had combined radium-226 and radium-228 concentrations greater than one-half the MCL (2.82 and 3.71 pCi/L, respectively). Combined radium-226 and radium-228 concentrations from this study appear elevated in comparison to a set of 1,421 samples from 1,368 wells sampled between 1992 and 2011 within 42 states for the NAWQA program. These NAWQA samples had combined radium-226 and radium-228 concentrations ranging from less than the ssLc to 56.0 pCi/L, with a median of 0.60 pCi/L (U.S. Geological Survey, 2012b); about 5 percent of the samples exceeded the MCL. Only one of the NAWQA samples was higher than the highest value measured in this study (39.1 pCi/L).

Polonium-210

Polonium-210 activities ranged from less than the ssLc to 3.08 pCi/L, with a median of 0.032 pCi/L (table 5, appendix 6). Twelve of the 40 sampled wells had values less than the ssLc, and all but 14 samples had values of less than 0.10 pCi/L. Three wells were considered elevated with polonium-210 activities greater than 1.0 pCi/L (one well completed in Ts and two wells completed in TKi). The U.S. Environmental Protection Agency currently (2013) does not have a MCL for polonium-210, and the only applicable USEPA drinking-water standard for polonium-210 is the adjusted gross alpha-particle activity standard of 15 pCi/L. Of the 27 wells that had polonium-210 activities considered detections (26 of which were wells completed in TKi rocks), all but 1 had gross alpha-particle activities greater than 15 pCi/L (range of 12.6 to 740 pCi/L, with a median of 40 pCi/L).

Hydrogeochemistry of Radiochemical Constituents in Groundwater

The geology and composition of aquifer materials as well as the geochemical, physical, and biological processes that can affect groundwater quality are factors controlling the occurrence and concentration of radiochemical constituents in natural waters. The importance of these various potential factors on radionuclide occurrence in the study area are investigated here by examining correlations among geology, concentrations of radionuclides and major ions, and other factors such as dissolved oxygen and pH. This section presents (1) general conclusions of this analysis and (2) more detailed analysis for each radiochemical constituent.

The occurrence and concentration of radiochemical constituents has been determined in some studies to be correlated with pH, dissolved oxygen, total dissolved solids, and redox conditions (Ayotte and others, 2011; Szabo and others, 2012; Wanty and Schoen, 1991). However, correlations between these properties and radiochemical constituents were not consistent based on the data collected for this study (figs. 13–15). Elevated radiochemical constituents generally occurred with similar pH, dissolved oxygen, and specific conductance (an indicator of relative total dissolved solids) conditions as those with low concentrations. For example, samples that exceeded one (or more) radiochemical drinking water standard or screening level had near-neutral pH values generally ranging from 6.2 to 7.8, and these values were similar to the pH values measured in most waters sampled for this study (fig. 13). Similarly, samples that exceeded one (or more) radiochemical drinking water standard or screening level were found for nearly the entire range of measured dissolved oxygen concentrations (fig. 14). And finally, a distinct relation between increasing specific conductance values and concentrations of radiochemical constituents was not apparent in this dataset (fig. 15). However, all samples that exceeded the USEPA MCL for uranium (30 µg/L) or the USEPA screening level for gross beta-particle activity (50 pCi/L) had specific conductance values higher than the median of all samples collected of 462 microsiemens per centimeter µS/cm (fig. 15); three samples that exceeded the USEPA combined radium-226 and radium-228 MCL (5 pCi/L) had specific conductance values higher than the median of 462 µS/cm (appendixes 3 and 6). Samples that exceeded the proposed MCL for radon (4,000 pCi/L) and the screening level for gross alpha-particle activity (15 pCi/L) had specific conductance values that included almost the entire range of measured values.

Reduction/oxidation (redox) conditions of sampled waters were classified based on concentration thresholds of available dissolved oxygen, nitrate (NO_3^-), manganese (Mn^{2+}), iron (Fe^{2+}), and sulfate (SO_4^{2-}) using methods of McMahon and Chapelle (2008) and Jurgens and others (2009). Classifications of redox conditions for individual samples are included in appendix 3. Calculated redox conditions were overwhelmingly (80 percent of the 169 samples) classified as either oxic (66 samples) or predominantly oxic (69 samples had dissolved oxygen concentrations greater than or equal to 0.5 mg/L, but missing one or more of the five listed parameters). The remaining samples were classified as either mixed (17 samples that generally exhibited both oxic and anoxic redox processes), anoxic (6 samples), predominately anoxic (7 samples with dissolved oxygen concentrations less than 0.5 mg/L, but missing one or more of the five parameters listed above), or indeterminate (4 samples).

Potential relations between radioactive constituents and calculated redox conditions were examined, but no clear relations were observed with the available data. Samples from the relatively small groups classified as anoxic or mixed had similar distributions and median concentrations of radioactive constituents (namely uranium and radon) as those within the predominant group of samples classified as oxic. Redox conditions in groundwater commonly are not in redox equilibrium and multiple redox conditions may exist simultaneously as water progresses from more oxygenated states to more reduced states (Jurgens and others, 2009). In addition, groundwater samples are often mixtures of water from multiple parts of an aquifer that may have different redox conditions. Additional data, such as the analysis of samples for speciated forms of key elements such as manganese and iron, are needed to further define redox processes and potential relations between redox conditions and radioactive constituents.

To further explore the relation between various groundwater constituents, correlations between radiochemical constituents and inorganic constituents and physical properties (table 6) and correlations among radiochemical constituents (table 7) were determined using the nonparametric Spearman statistical tests. Statistical tests were done using all data collected for this study, as well as subsets of the data from wells completed in the Boulder batholith and similar units (TKi). The analytical results from wells sampled multiple times were included in this analysis because conditions may have varied between sampling events. Statistically significant correlations described below are defined as strong (greater than or equal to +0.9 to +1.00 or less than or equal to -0.90 to -1.00), moderate (from +0.50 to +0.89 or from -0.50 to -0.89), and weakly or not correlating (from -0.49 to +0.49). Correlations are evaluated as to the degrees of significance (p) and reported with the correlation value.

Correlations between most radiochemical constituents and either major ions or physical properties were weakly correlated or not statistically significant (p-value >0.05). The strongest correlations, although considered moderate, were between potassium and gross beta-particle activities (rho ranged from 0.72 to 0.82), between total dissolved solids (as well as specific conductance, which is strongly correlated with total dissolved solids) and gross alpha-particle and gross beta-particle activities (rho ranged from 0.63 to 0.71), and between manganese and radium isotopes (rho ranged from 0.59 to 0.64) (table 6).

Evaluation of correlations of radiochemical constituents of all sampled wells indicated that moderate to strong correlations occurred between the radium isotopes (rho = 0.78 to 0.92) and between the gross alpha-particle and beta-particle activities (rho = 0.77 to 0.96) (table 7). These strong correlations are expected given most constituents are part of the same decay chain (for example, radium-224 and radium-228) or for the same constituent analyzed at different times or by different methods. In addition, these same correlations were strong (rho >0.90) for the subset of data for wells only completed in the TKi geologic unit. A strong correlation of radium-226 analyzed by radon emanation and by alpha spectrometry indicates good agreement between the analytical methods (rho = 0.92) (table 7).

Correlations between most constituents that are not part of same decay chain were not statistically significant (p-value >0.05) or were weakly to moderately correlated (table 7).

Figure 13.   Relation between pH and radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Figure 14.   Relation between dissolved oxygen and radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

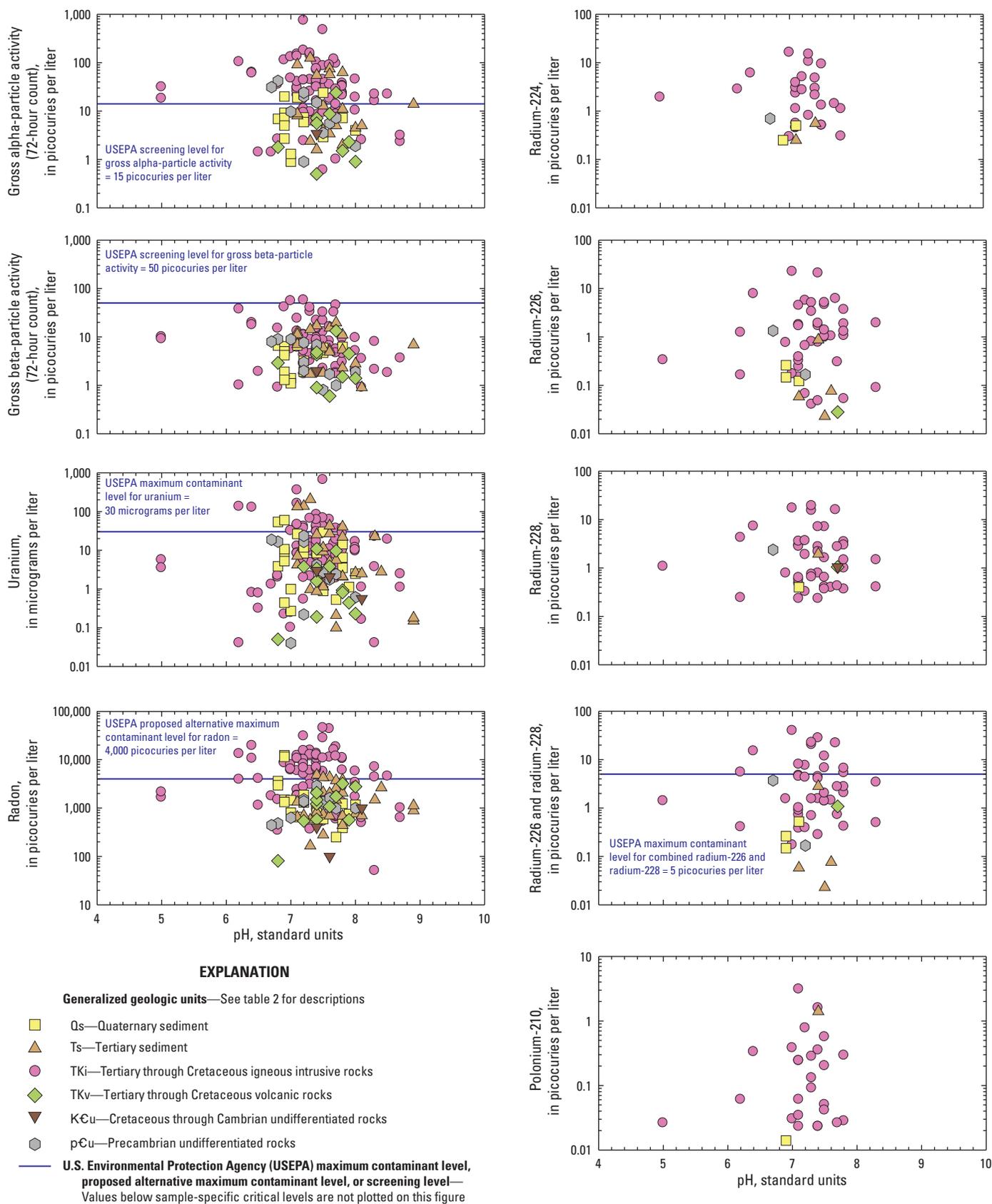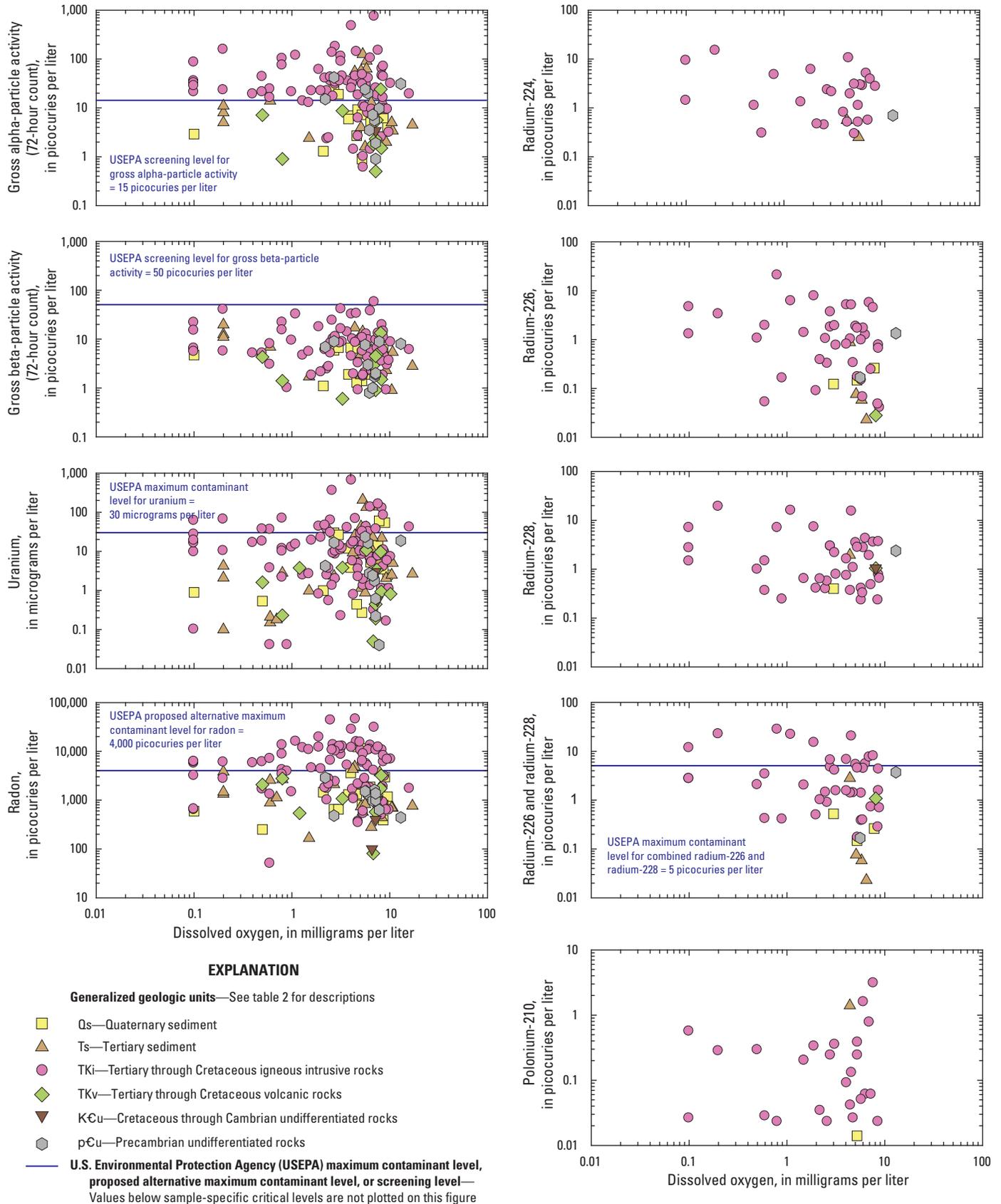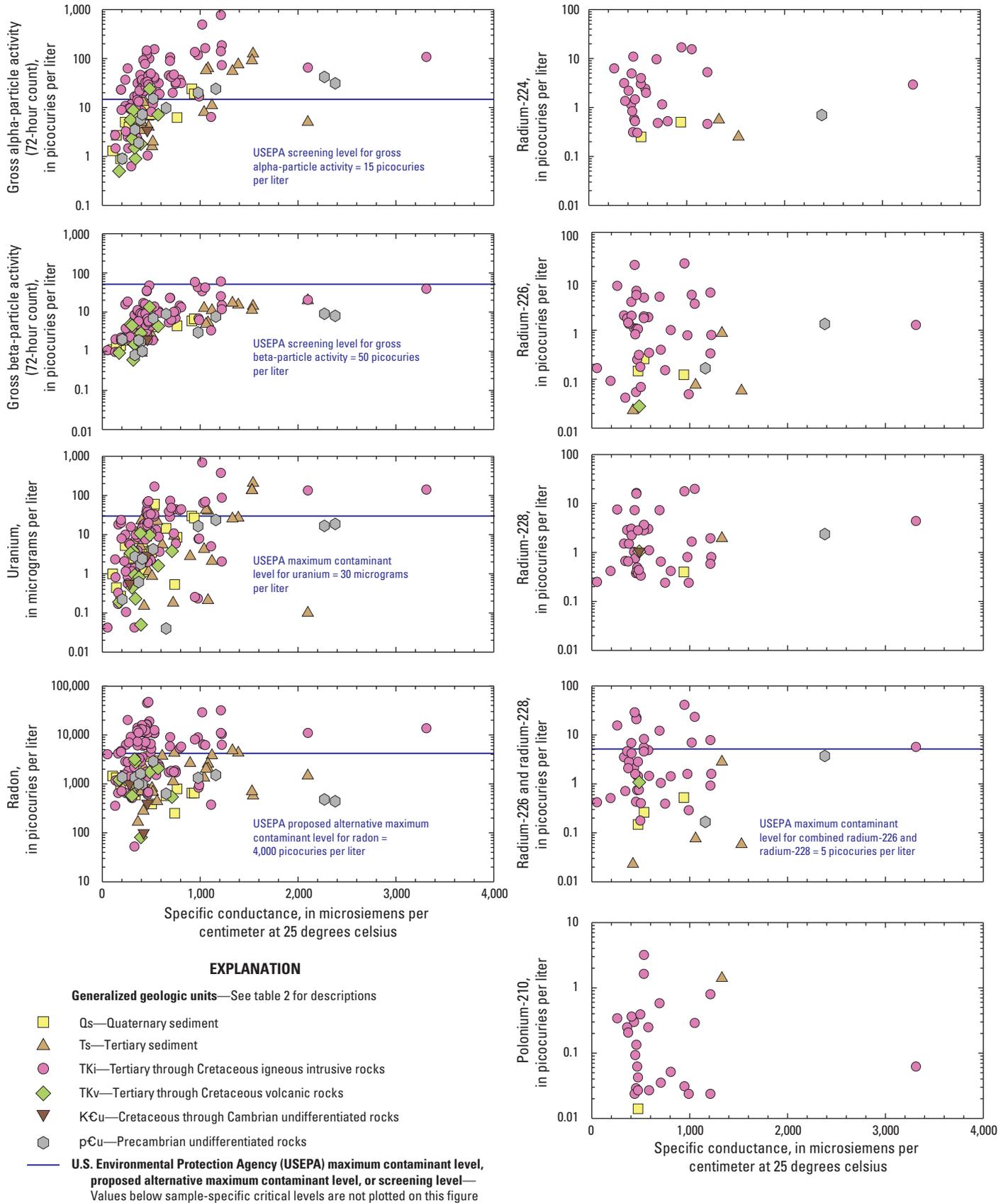**Figure 15.** Relation between specific conductance and radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Table 6. Spearman rank correlation coefficients between radiochemical constituents and major ions and physical properties of groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

[All coefficients shown are significant to the 95-percent confidence level (significant at p-value 0.05 or less). NS, correlation coefficient is not significant at the 95-percent confidence interval. Values in parentheses indicate number of samples]

Analyte (sample count)	Dissolved oxygen	pH, onsite	Specific conductance	Dissolved solids, sum of constituents	Hardness, as calcium carbonate	Calcium	Magnesium	Potassium	Sodium
Gross alpha-particle activity, 72-hour count	-0.27 (136)	NS (137)	0.62 (137)	0.63 (134)	0.51 (134)	0.44 (134)	0.52 (134)	0.54 (134)	0.46 (134)
Gross alpha-particle activity, 30-day count	-0.21 (136)	NS (137)	0.63 (137)	0.63 (134)	0.48 (134)	0.41 (134)	0.48 (134)	0.52 (134)	0.49 (134)
Gross beta-particle activity, 72-hour count	-0.25 (136)	-0.22 (137)	0.65 (137)	0.69 (134)	0.49 (134)	0.48 (134)	0.41 (134)	0.82 (134)	0.53 (134)
Gross beta-particle activity, 30-day count	-0.22 (136)	-0.18 (137)	0.70 (137)	0.71 (134)	0.55 (134)	0.51 (134)	0.49 (134)	0.72 (134)	0.54 (134)
Uranium, natural	NS (165)	NS (167)	0.50 (167)	0.48 (165)	0.55 (165)	0.47 (165)	0.58 (165)	0.36 (165)	0.28 (165)
Radium-224	NS (37)	NS (38)	NS (38)	NS (36)	NS (36)	NS (36)	NS (36)	NS (36)	NS (36)
Radium-226, alpha spectrometry method	NS (38)	NS (39)	NS (39)	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)
Radium-226, radon emanation method	-0.30 (50)	NS (51)	NS (51)	NS (49)	NS (49)	NS (49)	NS (49)	NS (49)	NS (49)
Radium-228	-0.28 (51)	NS (52)	NS (52)	NS (50)	NS (50)	NS (50)	NS (50)	NS (50)	NS (50)
Radon-222	-0.27 (161)	-0.19 (163)	NS (163)	NS (163)	NS (163)	NS (163)	NS (163)	0.26 (163)	NS (163)
Polonium-210	NS (38)	NS (39)	NS (39)	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)

Analyte (sample count)	Alkalinity, onsite	Chloride	Fluoride	Silica	Sulfate	Nitrate plus nitrite, as nitrogen	Iron	Manganese
Gross alpha-particle activity, 72-hour count	0.49 (128)	0.49 (134)	0.33 (106)	NS (134)	0.49 (134)	NS (103)	0.22 (134)	0.37 (121)
Gross alpha-particle activity, 30-day count	0.50 (128)	0.50 (134)	0.32 (106)	0.17 (134)	0.53 (134)	0.20 (103)	NS (134)	0.26 (121)
Gross beta-particle activity, 72-hour count	0.45 (128)	0.52 (134)	0.45 (106)	0.42 (134)	0.60 (134)	NS (103)	0.33 (134)	0.49 (121)
Gross beta-particle activity, 30-day count	0.54 (128)	0.56 (134)	0.40 (106)	0.27 (134)	0.58 (134)	NS (103)	NS (134)	0.38 (121)
Uranium, natural	0.45 (159)	0.45 (165)	NS (136)	NS (165)	0.28 (165)	0.38 (132)	NS (165)	NS (125)
Radium-224	NS (36)	NS (36)	NS (36)	NS (36)	NS (36)	NS (6)	0.51 (36)	0.62 (36)
Radium-226, alpha spectrometry method	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)	NS (6)	0.50 (37)	0.59 (37)
Radium-226, radon emanation method	NS (49)	NS (49)	NS (49)	NS (49)	NS (49)	NS (19)	0.54 (49)	0.61 (37)
Radium-228	NS (50)	NS (50)	NS (50)	NS (50)	NS (50)	NS (19)	0.52 (50)	0.64 (38)
Radon-222	NS (157)	NS (163)	NS (134)	NS (163)	NS (163)	NS (129)	NS (163)	0.30 (124)
Polonium-210	NS (37)	NS (37)	NS (37)	NS (37)	NS (37)	NS (6)	NS (37)	0.45 (37)

Table 7. Spearman rank correlation coefficients for radiochemical constituents for groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

[All coefficients shown are significant to the 95-percent confidence level (significant at p-value of 0.05 or less). Numbers in bold represent strong correlation with coefficient of + 0.90 or greater. NA, not applicable; --, data shown elsewhere in table; NS, indicates that correlation coefficient is not significant at the 95-percent confidence interval. Values in parentheses indicate number of samples]

Analyte (sample count)	Gross alpha-particle activity, 72-hour count	Gross alpha-particle activity, 30-day count	Gross beta-particle activity, 72-hour count	Gross beta-particle activity, 30-day count	Uranium, natural	Radium-224	Radium-226, alpha spectrometry method	Radium-226, radon emanation method	Radium-228	Radon-222	Polonium-210
Gross alpha-particle activity, 72-hour count	NA	**0.96 (138)**	0.82 (138)	**0.91 (138)**	0.75 (136)	0.56 (39)	0.58 (40)	0.59 (52)	0.55 (53)	0.60 (134)	0.41 (40)
Gross alpha-particle activity, 30-day count	--	NA	0.77 (138)	**0.90 (138)**	0.78 (136)	NS (39)	NS (40)	0.44 (52)	0.36 (53)	0.58 (134)	NS (40)
Gross beta-particle activity, 72-hour count	--	--	NA	0.89 (138)	0.49 (136)	0.68 (39)	0.68 (40)	0.63 (52)	0.64 (53)	0.51 (134)	0.42 (40)
Gross beta-particle activity, 30-day count	--	--	--	NA	0.73 (136)	0.36 (39)	0.43 (40)	0.44 (52)	0.32 (53)	0.57 (134)	0.32 (40)
Uranium, natural	--	--	--	--	NA	NS (38)	NS (39)	NS (51)	NS (52)	0.39 (163)	NS (39)
Radium-224	--	--	--	--	--	NA	**0.91 (39)**	0.83 (38)	**0.92 (39)**	0.51 (36)	0.57 (39)
Radium-226, alpha spectrometry method	--	--	--	--	--	--	NA	**0.92 (39)**	0.84 (40)	0.55 (37)	0.61 (40)
Radium-226, radon emanation method	--	--	--	--	--	--	--	NA	0.78 (52)	0.61 (49)	0.61 (39)
Radium-228	--	--	--	--	--	--	--	--	NA	0.36 (50)	0.49 (40)
Radon-222	--	--	--	--	--	--	--	--	--	NA	0.56 (37)
Polonium-210	--	--	--	--	--	--	--	--	--	--	NA

In addition, relations between several radiochemical constituents of the same decay chain also were not statistically significant or were weakly correlated. These poor correlations indicate that concentrations of radiochemical constituents were affected not only by the presence of parent radionuclides but also by other geochemical processes that can affect the occurrence and mobility of these constituents. For example, uranium, radium-226, and radon are part of the uranium-238 decay series, but correlations among them were weak or not statistically significant. Also, some constituents were moderately to strongly correlated even though they do not originate from the same decay chain, suggesting multiple original elemental sources of the radiochemical constituents. For example, concentrations of radium isotopes were moderately to strongly correlated (rho = 0.78 to 0.92) likely because all are mobile under the same chemical conditions (Zapecza and Szabo, 1988) and can be mobilized as a result of radionuclide recoil effects (Fleischer, 1980).

Uranium

Ayotte and others (2011) reported that uranium in groundwater samples collected nationwide generally occurred most frequently in samples with oxic or mixed redox conditions and neutral to alkaline pH. Uranium generally is more soluble in oxidizing conditions (Wanty and Schoen, 1991). Although dissolved oxygen alone is generally not a good measure of the redox state in groundwater systems, Morrow (2001) determined that uranium concentrations of 1.0 µg/L or more were more frequent in samples where dissolved-oxygen concentrations were greater than 1 mg/L. Samples with elevated uranium concentrations from this study were reported for nearly the entire range of measured pH, specific conductance, and dissolved oxygen conditions (figs. 13 to 15). Uranium was not correlated with dissolved oxygen (p-value = 0.78, rho = 0.02); concentrations of uranium exceeded the 30 µg/L MCL in 31 of 168 samples with dissolved oxygen concentrations ranging from less than 0.2 to greater than 10.0 mg/L (fig. 14). Uranium solubility can increase with increasing pH over the normal pH range of most waters. Although uranium was not correlated with pH (p-value = 0.28, rho = -0.08), 27 of 31 samples that exceed the uranium MCL had pH concentrations of 7 or higher (fig. 13). Uranium concentrations were weakly correlated with specific conductance (rho = 0.50); however, concentrations greater than the MCL occurred only in samples with specific conductance values higher than the median value of 462 µS/cm (fig. 13, table 6). Uranium was only weakly to moderately correlated with major ions; the strongest correlations were with hardness (rho = 0.55) and magnesium (rho = 0.58) (table 6).

The strongest correlations between uranium and other radiochemical constituents were with gross-alpha-particle activity, gross beta-particle activity, and radon (table 7, fig. 16). Although the highest uranium concentrations typically were associated with some of the highest radon concentrations

and gross alpha- and beta-particle activities, these latter constituents also were elevated in some samples with relatively low uranium concentrations. Correlations between uranium and other radiochemical constituents (including the radium isotopes) were not significant (p-value >0.05).
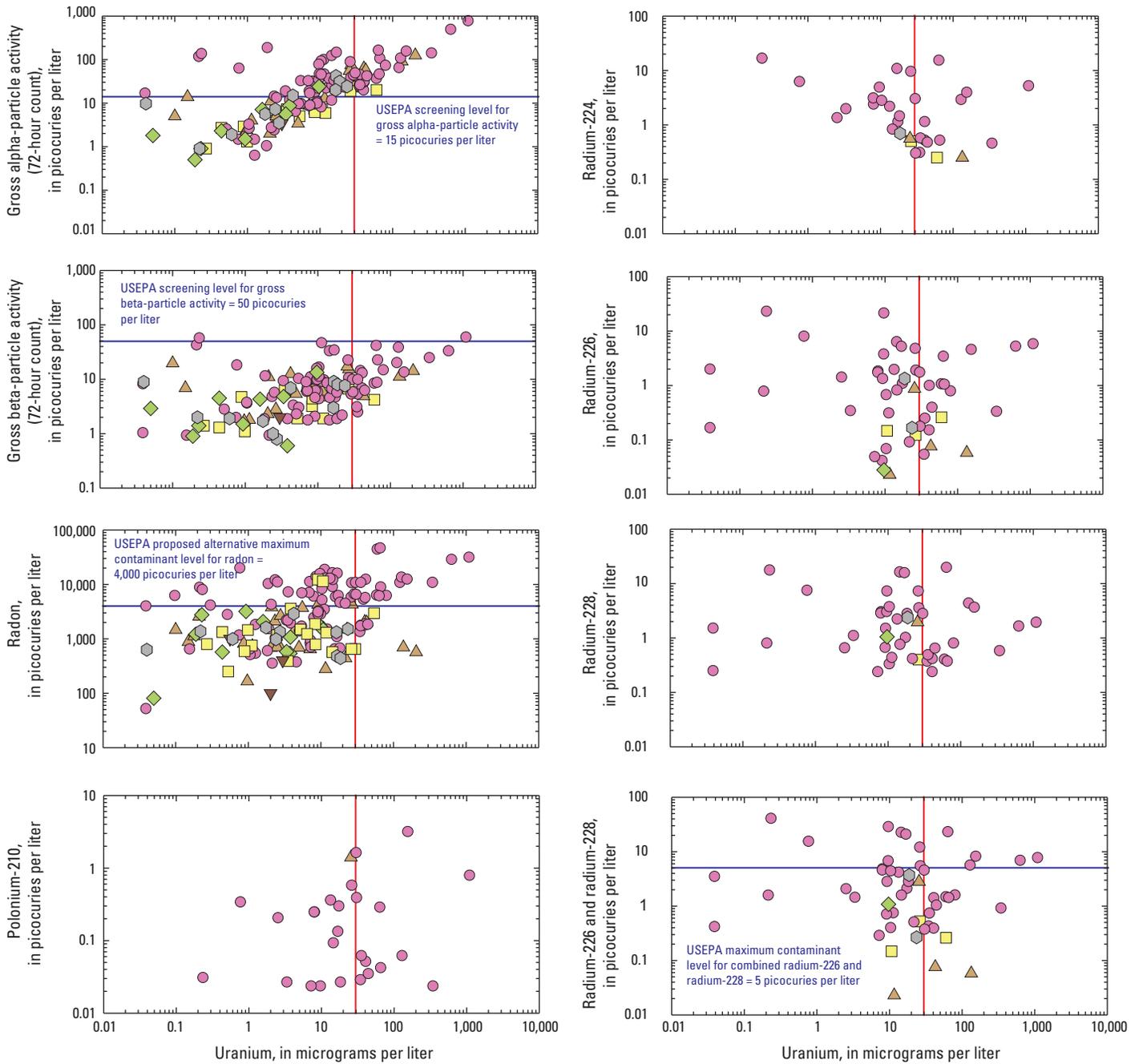
Radon

Radon was elevated in nearly all groundwater samples from the TKi geologic unit presumably owing to the abundant uranium-bearing minerals in those rocks (Tilling and Gottfried, 1969). As a noble gas, radon is not ionized in solution and does not precipitate in solid phases. Although it can be adsorbed to organic material, radon typically is not adsorbed to other surfaces (Wanty and Schoen, 1991). Because of these properties, radon typically is elevated in areas that are enriched in its parent elements.

Ayotte and others (2011) reported that the occurrence of radon in groundwater samples collected nationwide occurred most commonly in oxic water with low pH. However, samples with elevated radon concentrations from this study occurred over nearly the entire range of measured pH, specific conductance, and dissolved oxygen conditions (figs. 13 to 15). Radon was only weakly correlated with a few major ions and physical properties including dissolved oxygen, pH, manganese, and potassium (table 6). The correlation between radon and radiochemical constituents was generally stronger, with moderate correlations with gross alpha-particle activity, gross beta-particle activity, radium-224, and radium-226; radon was only weakly correlated with uranium and radium-228 (table 7). Similar correlations existed for the samples from the wells completed in the TKi geologic unit. The highest radon concentrations typically were associated with some of the highest uranium concentrations and alpha-particle activities (fig. 17).

Gross Alpha-Particle Activity

Samples with measureable concentrations of gross alpha-particle activities occurred over nearly the entire range of pH, specific conductance, and dissolved oxygen conditions measured during the study (figs. 13 to 15). Gross alpha-particle activity (72-hour count) was moderately correlated (rho ranging from 0.51 to 0.63) with specific conductance, total dissolved solids, hardness, magnesium, and potassium (table 6). Gross alpha-particle activity (30-day count) was moderately correlated (rho ranging from 0.50 to 0.63) with specific conductance, total dissolved solids, potassium, alkalinity, chloride, and sulfate (table 6). The strongest correlations were with specific conductance and total dissolved solids, suggesting the association of increased gross alpha-particle activity with increased dissolved solids. Most samples (54 of 70 samples, 77 percent) with specific conductance values greater than the overall median value of 462 µS/cm had gross alpha-particle activities greater than the 15 pCi/L screening level (fig. 15).

Figure 16.   Relation between uranium and other radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Figure 17.   Relation between radon and other radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

The correlations between gross alpha-particle activities (72- hour and 30-day counts) and between gross alpha-particle activities (72- hour and 30-day counts) and gross beta-particle activity (30-day count) were strong (rho >0.90) (table 7). Gross alpha-particle activity (72-hour count) was moderately correlated (rho = 0.55 to 0.75) to all other radioactive constituents except polonium-210 (which was only weakly correlated, rho = 0.41). Gross alpha-particle activity (30-day count) was moderately correlated with uranium and radon (rho = 0.78 and 0.58, respectively) (table 7). Similar correlations existed for the samples from the wells completed in the TKi geologic unit. Elevated gross alpha-particle activities were associated with some of the highest uranium and radon concentrations and gross beta-particle activities (fig. 18).

Gross Beta-Particle Activity

Samples with measureable gross beta-particle activities occurred over nearly the entire range of measured pH, specific conductance, and dissolved oxygen (figs. 13 to 15). Gross beta-particle activity (72-hour count, 30-day count, or both) was moderately correlated (defined as rho values of 0.50 to 0.89) with specific conductance, total dissolved solids, hardness, calcium, potassium, sodium, alkalinity, chloride, and sulfate (table 6). The correlations with specific conductance and total dissolved solids (table 6) suggest that increased gross beta-particle activity is associated with increased dissolved solids. For example, all samples that exceeded the gross beta-particle activity screening level of 50 pCi/L were associated with specific conductance values greater than the overall median of 462 µS/cm (fig. 15).

The strongest correlation between gross beta-particle activity and major ions was with potassium (table 6). Gross beta-particle activities typically increased with increased potassium concentrations (fig. 19), likely because one potassium isotope (potassium-40) is a beta-particle emitter. Potassium-40 is a likely source of gross beta-particle activity in groundwater because potassium is a relatively common element, comprising about 2.6 percent of the Earth's crust (Fleischer, 1953) and even larger proportions in granitic rocks (such as the TKi geologic unit), which contain potassium-rich minerals such as potassium feldspar. Aerial radiometric surveys of the study area show that much of the area underlain by the TKi geologic unit had elevated potassium concentrations in the upper 50 centimeters of surface soil and rock relative to surrounding areas (Duval and others, 2005). Tilling and Gottfried (1969) reported average potassium concentrations of 3.3 percent for samples of the Boulder batholith.

Gross beta-particle activity was correlated with other radiochemical constituents (table 7). The strongest correlations were between gross beta-particle and gross alpha-particle activities (rho ranging from 0.77 to 0.91) and between gross beta-particle activity counted at 72 hours and 30 days (rho = 0.89). Gross beta-particle activity (72-hour count) was moderately correlated to the radium isotopes (rho ranging from

0.63 to 0.68) as well as radon (rho = 0.51). Gross beta-particle activity (30-day count) was moderately correlated only with uranium and radon (rho = 0.73 and 0.57, respectively; table 7). Similar correlations existed for the samples from the wells completed in the TKi geologic unit. The highest gross beta-particle activities typically were associated with the highest gross alpha-particle activities and some of the highest uranium, radon, and radium concentrations (fig. 20).

Radium

Geochemical conditions commonly associated with elevated concentrations of radium in groundwater include (1) low levels of dissolved oxygen (dissolved oxygen less than 1 milligram per liter), (2) acidic water (pH less than 6), and (3) water with high concentrations of dissolved solids (especially calcium, barium, magnesium, strontium, potassium, sulfate, or bicarbonate) (Szabo and others, 2012). However, samples with elevated concentrations of radium isotopes from this study occurred over nearly the entire range of dissolved oxygen, pH, and specific conductance conditions (figs. 13–15).

Radium isotopes were not correlated significantly (p-value >0.05) with pH, specific conductance, total dissolved solids, and most other major ions (table 6). However, all but one sample that exceeded the combined radium-226 and radium-228 MCL of 5 pCi/L were associated with specific conductance values higher than the median of 462 µS/cm. The correlations between dissolved oxygen and radium-226 or radium-228 isotopes were only weak or not significant. Radium isotopes were moderately correlated with iron and manganese (rho ranging from 0.50 to 0.64).

Radium isotopes were moderately to strongly correlated to one another (table 7), probably because all radium isotopes are mobile under the same chemical conditions (Zapecza and Szabo, 1988) and all can be mobilized as a result of radionuclide recoil effects (Fleischer, 1980). The general association among the radium isotopes is displayed in figure 21. The general co-occurrence of radium-224 and radium-228 is apparent as many data plot near the 1:1 line (fig. 21*A*). Focazio and others (2001) reported that radium-224 and radium-228 co-occur in nearly equal concentrations in samples from a variety of geohydrologic settings throughout the Nation. The high degree of correlation between radium-224 and radium-228 (table 7) is predictable as these isotopes originate from the same decay series. The weaker correlation between radium-226 and radium-228, as well as radium-224 and radium-226, is apparent with many samples not plotting near the 1:1 line (figs. 21*B* and 21*C*).

Although there is no current (2013) MCL for radium-224, Parsa (1998) calculated that radium-224 could contribute considerable gross alpha-particle activity to a water sample for about 7 days after sample collection. Because radium-224 was analyzed within 96 hours of sample collection for this study, the measurable occurrence of radium-224 may support at least part of the observed decrease in gross alpha-particle activity
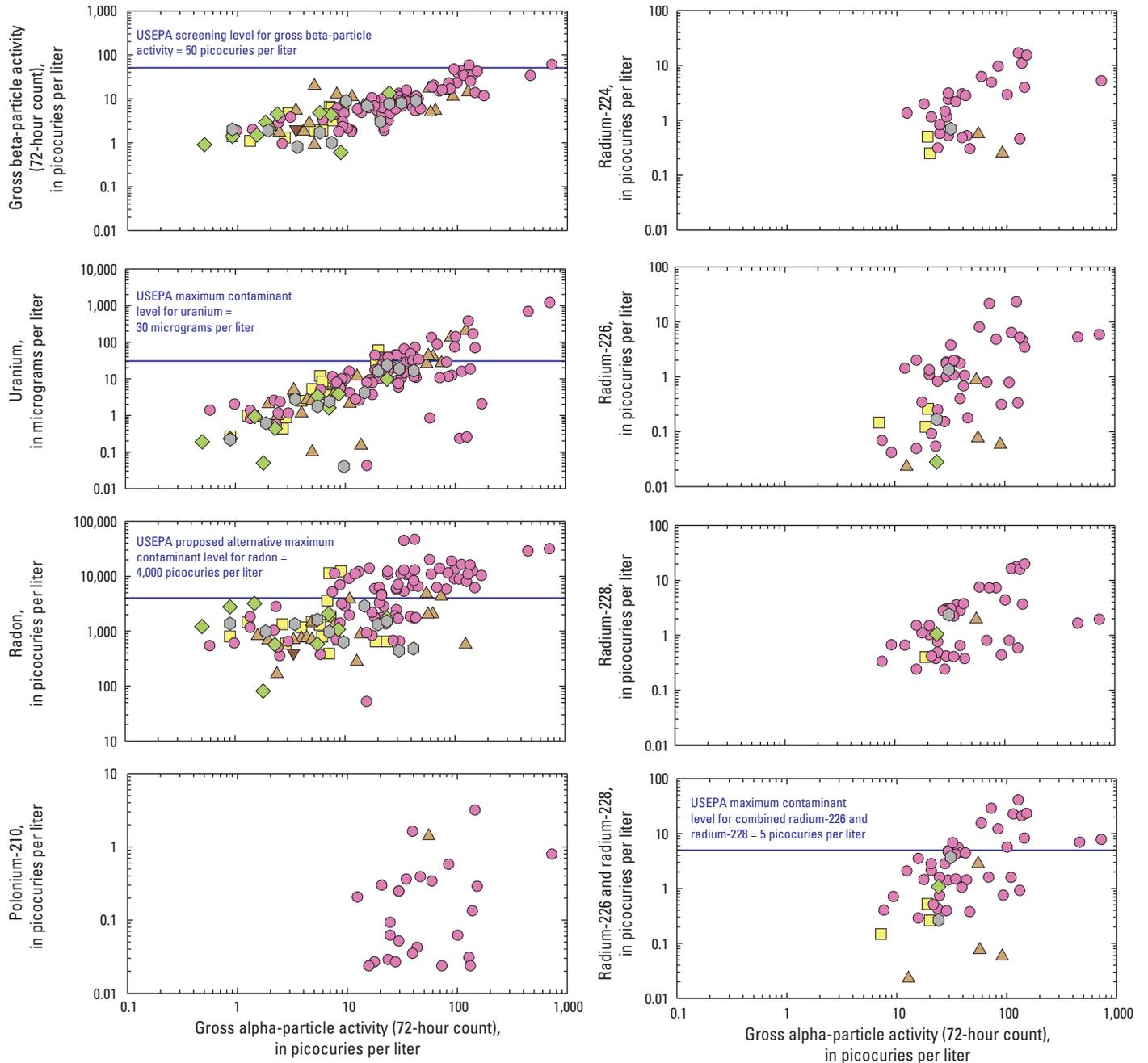
Figure 18.   Relation between gross alpha-particle activity (72-hour count) and other radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Figure 19. Relation between potassium and gross beta-particle activity measured in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. *A*, 72 hours after sample collection. *B*, 30 days after sample collection.

Figure 20.   Relation between gross beta-particle activity (72-hour count) and other radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.
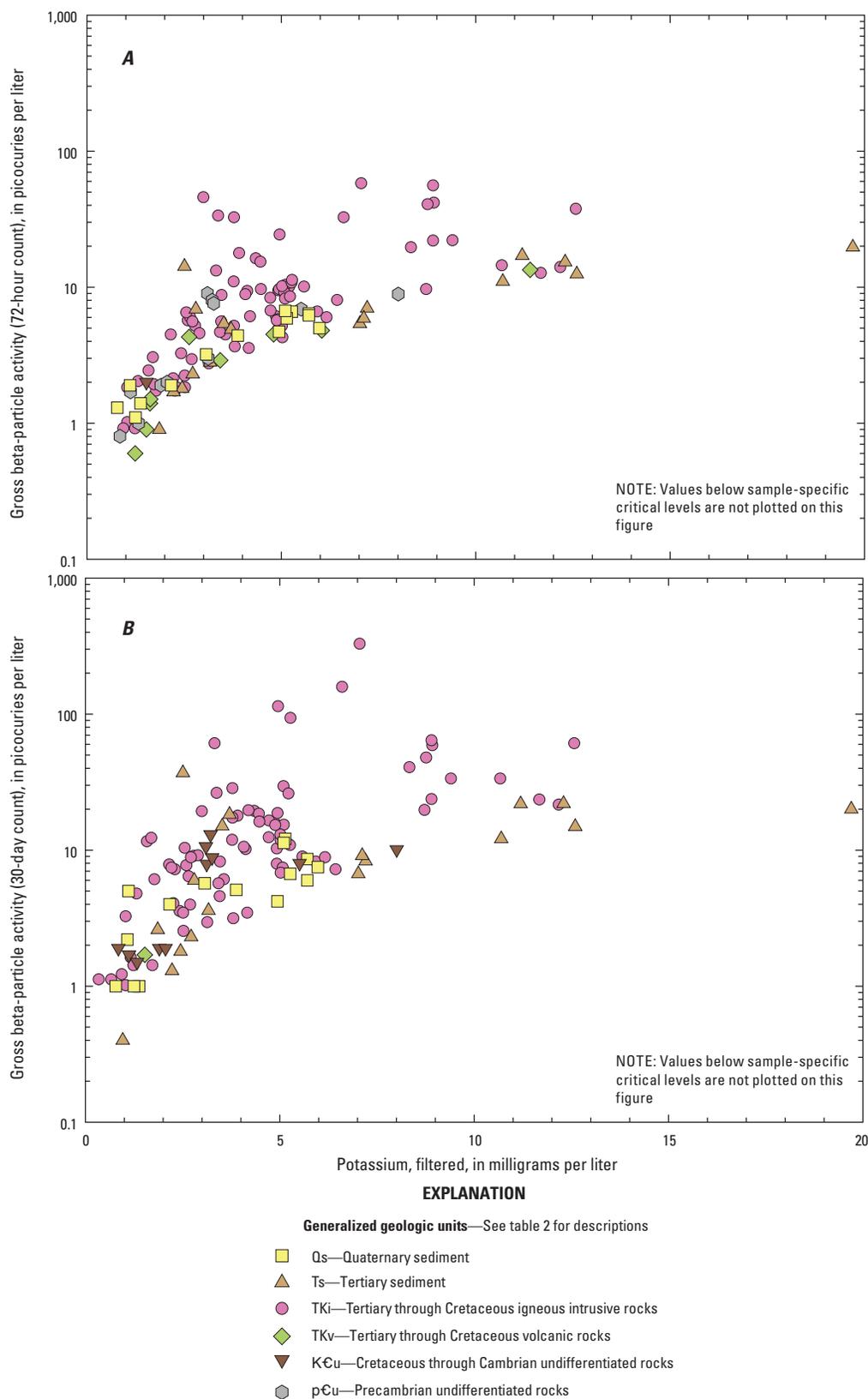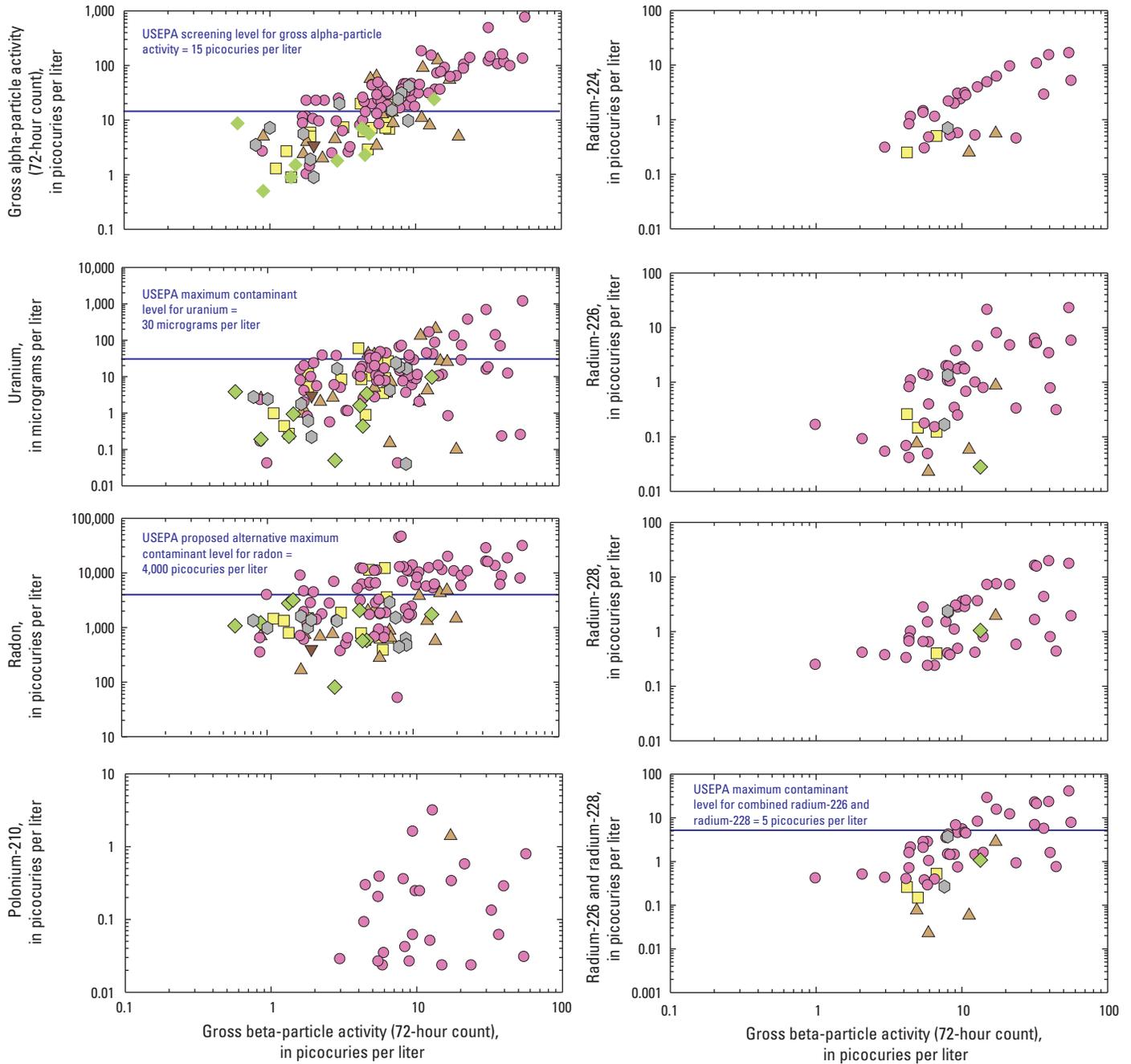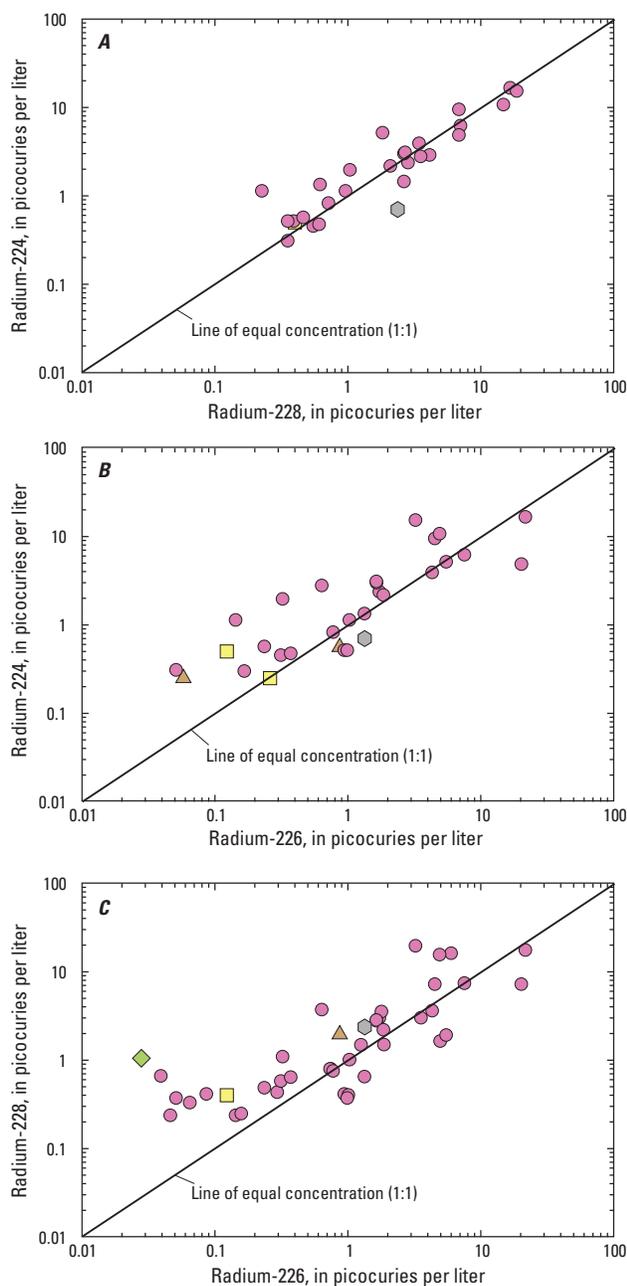
NOTE: Values below sample-specific critical levels are not plotted on this figure

EXPLANATION

Generalized geologic units—See table 2 for descriptions

- ☐ Qs—Quaternary sediment
- △ Ts—Tertiary sediment
- ⬤ TKi—Tertiary through Cretaceous igneous intrusive rocks
- ◇ TKv—Tertiary through Cretaceous volcanic rocks
- ⬡ p€u—Precambrian undifferentiated rocks

Figure 21. Relations between selected radiochemical constituents in groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010. *A*, radium-224 and radium-228; *B*, radium-224 and radium-226; and *C*, radium-228 and radium-226.

with time in some samples. A median 53 percent decrease was observed between the 72-hour and 30-day gross alpha-particle activity counts in samples from 19 wells with radium-224 values of 1.0 pCi/L or more.

Polonium-210

Polonium-210 concentrations were typically low and were less than 0.10 pCi/L in 26 out of 40 samples (13 samples were considered nondetects). Samples with measureable concentrations of polonium-210 occurred over nearly the entire range of pH, specific conductance, and dissolved oxygen conditions (figs. 13 to 15). Samples with polonium-210 concentrations greater than 1.0 pCi/L were from the Ts and TKi units and had neutral pH values (7.0 to 7.4), relatively high dissolved oxygen conditions (greater than 4.0 mg/L), and specific conductance values higher than the study median value of 462 µS/cm. Polonium-210 also was not significantly correlated with physical properties, major ions, or iron, and only weakly correlated with manganese (rho = 0.45, table 6).

The relation of polonium-210 with other radiochemical constituents indicated that the strongest correlations, although considered moderate (rho = 0.61 to 0.56), were with radium-226, radium-224 and radon (table 7). Polonium-210 was not significantly correlated with uranium (*p*-value >0.05). Interestingly, polonium-210 had the weakest correlations (ranging from not statistically significant to rho = 0.42) with gross alpha-particle activity and gross beta-particle activity in comparison to the other radiochemical constituents.

Temporal Variation of Radionuclides

Thirty-eight wells were sampled twice during this study to examine temporal variability of uranium and radon concentrations as well as gross alpha-particle and beta-particle activities. Two of the wells were sampled 3 times. The time period between successive sampling events ranged from about 1 to 10 months for 29 wells to about 3 years for the other 9 wells. Variability between samples was evaluated using relative percent difference (RPD), which was calculated by dividing the difference of the two values by the mean of the two values and reporting the absolute value of this quotient as a percent. Because some variability would be expected due to sampling and analytical error, a RPD larger than 30 percent was chosen as an indicator of actual temporal change in concentration or activity.

Radiochemical constituents measured in most resampled wells indicated limited temporal variability (RPDs <30 percent). However, some resampled wells did report relatively large temporal variation (RPDs >30 percent) of individual radiochemical constituents. Temporal variation of gross alpha-particle activities with RPDs larger than 30 percent was observed among 38 percent of the resampled wells. Other constituents that had large temporal variability (RPDs >30 percent) among resampled wells include: uranium (29 percent of the resampled wells), gross-beta particle activity (15 percent of

the resampled wells), and radon (11 percent of the resampled wells). These large temporal variations occurred at all sampling intervals, whether it was 1–2 months, 10 months, or 3 years.

Some temporal variations were large and occurred throughout the concentration range of most samples. For instance, uranium concentrations measured in 4 of the 38 resampled wells had RPDs of 155–198 percent. These variations occurred at low concentrations (0.10 µg/L to 0.79 µg/L in well 462813112060701), moderate concentrations (for example, 10.1 µg/L to 1.31 µg/L in well 460143112331701), and high concentrations (1.94 µg/L to 354 µg/L and 9.89 µg/L to 159 µg/L in wells 461260112050501 and 462555111580601, respectively). For the latter two wells, uranium concentration increased from a value less than the 30 µg/L MCL in the first sample, to a value greater than the MCL in the second sample.

Temporal variability in water quality was observed in a set of wells sampled on more than one occasion, suggesting the need for multiple samples collected over a range of conditions throughout time to adequately evaluate water-quality conditions. Radionuclide concentrations in groundwater can change for a variety of reasons. Changes in groundwater level or flow direction can cause groundwater from different sources to flow to the well. These different sources may have different radionuclide concentrations because of (1) differences in geochemical characteristics, such as varying redox condition or bicarbonate concentration affecting radionuclide mobility, or (2) differences in the amount of source material in the aquifer materials. Insufficient data are available to provide definitive explanations for the temporal variations observed in this investigation.

Implications of Radionuclide Occurrence

Waters sampled for this study were considered elevated in radionuclides relative to national values collected for the USGS National Water Quality Assessment program. Granitic rocks, such as the TKi geologic unit of this study, are known sources of elevated uranium (Tilling and Gottfried, 1969). Although concentrations of radiochemical constituents in samples from wells completed in this unit were typically among the most elevated in this study, commonly exceeding drinking-water standards, the water quality in several TKi wells was acceptable. Although nearly one-half of the samples analyzed were collected from the TKi, elevated radiochemical constituents were observed in five of the six generalized geologic units examined. Only wells with measured or suspected elevated concentrations of uranium or radon, or elevated activities of gross alpha-particle or gross beta-particle, were sampled for the more extensive set of radionuclides (radium isotopes, uranium isotopes, and polonium-210). Because most samples analyzed for the more extensive list of radionuclides were from wells completed in TKi, evaluation of these constituents in other geologic units is limited. Therefore, further areal evaluation of other generalized geologic units is warranted.

Although several reports describe relations between the occurrence and concentrations of radiochemical constituents and pH, dissolved oxygen, total dissolved solids, and major-ion chemistry, these do not appear to be dominant factors controlling the presence and concentration of radiochemical constituents in the study area. Elevated radiochemical constituents generally were measured in samples with similar pH, dissolved oxygen, and specific conductance (an indicator of relative total dissolved solids) as those with low concentrations. Interestingly, constituents within the same radioactive decay series were not necessarily strongly correlated suggesting that geochemical processes beyond the presence of the parent isotope controlled the occurrence and concentration of radiochemical constituents in these waters.

The data collected for this study indicate that, in general, predicting the occurrence of elevated radionuclide concentrations in water samples collected from wells in the study area has a high degree of uncertainty. Although wells completed in the Boulder batholith or similar geologic units were more likely to produce groundwater with elevated radionuclides, concentrations did vary, even in wells that were completed in the same geologic unit and were in close proximity to one another (less than 1,000 feet (ft) apart). The occurrence and mobility of radionuclides in groundwater are controlled by the presence of source radionuclides in the aquifer materials; hydrologic characteristics of the aquifers; and chemical and physical processes (precipitation, adsorption, and ion exchange) that occur as water flows through and interacts with the aquifer matrix. Hydrologic characteristics are important in explaining the differences in fracture-rock aquifers (such as TKi, TKv, and pꞜu) and sedimentary aquifers (such as Qs, Ts, and Ꞓ). Differences in the hydrogeologic setting of the flow system in these aquifer types (generalized in fig. 4) can substantially affect residence time, water/rock volumetric ratio, and the surface area of the rock per volume of water, all of which affect radionuclide occurrence in groundwater (Wanty and Schoen, 1991). Thus, wells in close proximity to each other sometimes produced water with different chemistry, likely because of differing residence times, dissimilar mineralogy, and varying geochemical conditions (such as pH, dissolved oxygen, and temperature) encountered along their distinct flow paths.

Data from this study illustrate that holding times before sample analysis can affect results, interpretation, and decisions regarding actions relative to water-quality criteria or concerns. Radiochemical constituents analyzed for this study typically were analyzed within 4 days or less of sample collection. With the exception of gross alpha- and beta-particle activities, all radionuclides were decay corrected (reported at the levels they would have been when the samples were collected). This correction is especially important for short-lived radionuclides such as radon-222, polonium-210, and radium-224 because the decay corrected values more adequately represent the levels of radionuclides consumed by the public using domestic wells as their source of drinking water.

Gross alpha- and beta-particle activities were counted at 72 hours and 30 days after sample collection. Gross alpha-particle activities typically decreased between the 72-hour and 30-day counts. This decreased activity with time indicates the presence of short-lived radionuclides (constituents with relatively short half-lives). Samples stored for long periods of time before analysis could result in the decay of short-lived radionuclides (such as radium-224) to levels considered as nondetects. These nondetects would not allow for the correction of concentrations to the time of sample collection as described previously. Gross beta-particle activity measurements commonly over-estimate the abundance of beta-emitting radionuclides present in groundwater. The data suggest that gross beta-particle activities could be affected by ingrowth of beta-emitting radionuclides during holding time before laboratory analysis as gross beta-particle activities typically increased between the 72-hour and 30-day counts.

Statistically significant correlations among radiochemical constituents were observed and could be used as screening tools for future assessments or to aid private well owners when assessing the water quality of their drinking-water supply. As in this study, Focazio and others (2001) reported that radium-224 and radium-228 co-occur in nearly equal concentrations in samples from a variety of geohydrologic settings throughout the Nation. Because of this approximate 1:1 relation, it may be reasonable to use radium-228 as a screening tool for the much shorter lived radium-224 isotope. For example, elevated radium-228 concentrations may indicate the need for additional radium-224 analysis. U.S. Environmental Protection Agency (2012c) reported that the occurrence of radionuclides in groundwater can be found by using gross alpha-particle activity as a test. If high levels of gross alpha-particle activity are reported, additional tests would be needed to identify which radionuclides are present (U.S. Environmental Protection Agency, 2012c). For this study, gross alpha-particle activity (72-hour count) appeared to be an effective screening tool for the identification of radiochemical constituents that exceeded USEPA drinking-water standards, proposed standards, or screening levels. All samples exceeding the USEPA MCL for uranium and combined radium-226 and radium-228 had gross alpha-particle activities (72-hour count) of 15 pCi/L or greater. It is important to understand that radium-228 is a beta-particle emitter and many studies have documented that the gross alpha-particle screen used as a first step to determine combined radium (radium-226 and radium-228) compliance could be inadequate in some settings (Focazio and others, 2001). All samples with radon concentrations greater than the proposed AMCL of 4,000 pCi/L had associated gross alpha-particle activities of 7 pCi/L or more. Similarly, all samples with elevated polonium-210 concentrations (1.0 pCi/L or greater) had associated gross alpha-particle activities of 40 pCi/L or greater. Interestingly, all samples that exceeded the 50 pCi/L screening level for gross beta-particle activity (30-day count) also had elevated gross alpha-particle activities (103 pCi/L or greater).

Public water-supply systems are required by both state (Montana Department of Environmental Quality, 2012) and federal (U.S. Environmental Protection Agency, 2009) agencies to have water analyzed for a list of constituents at regular intervals. No such requirements exist for private domestic wells and therefore, homeowners with a domestic well are responsible for evaluating the safety of their drinking water. Data from this study illustrate the importance of having the water from domestic wells evaluated for constituents that are potentially hazardous to human health. Because radionuclides as well as other constituents of human-health concern typically are detected only by laboratory analysis of water samples, individuals relying on domestic wells may unknowingly be exposed to elevated concentrations of one (or more) of these constituents.

This study also illustrates the importance of evaluating domestic drinking-water sources through time, as conditions and water chemistry did fluctuate in some wells. Temporal variability of uranium and radon concentrations, as well as gross alpha-particle and beta-particle activities, was observed for some wells based on two samples collected sequentially after time intervals ranging from about 1 month to 3 years. The uranium and radon concentration results suggest that sampling a well a few times over a range of hydrologic conditions may provide more representative radionuclide concentration data for a specific well.

Summary

Groundwater is the primary source of drinking water for most rural residents and municipalities of Jefferson County and surrounding areas in southwestern Montana. In response to concerns about radionuclides in groundwater, particularly the granitic rocks of the Boulder batholith, and the increasing reliance on groundwater for drinking water in this area, the U.S. Geological Survey (USGS), in cooperation with Jefferson County and the Jefferson Valley Conservation District, initiated a multi-year study to assess groundwater quality. During the study, water-quality data from existing wells were collected to determine the distribution of radioactive elements in groundwater in relation to well location, hydrogeologic unit, and geochemical conditions conducive to mobilization of these elements.

Scientists from the USGS collected a total of 168 samples from 128 wells within Jefferson County and six neighboring counties. Most study wells were the primary water supplies for private residences. Samples were collected from wells with well screens or open intervals within one of six generalized geologic units and analyzed primarily for uranium, radon, gross alpha-particle activity, and gross beta-particle activity. Wells with elevated concentrations or activities were sampled a second time to examine temporal variability in water quality, and these samples were analyzed for an expanded list of radioactive constituents including polonium-210, three isotopes of

radium, and three isotopes of uranium. A few wells located in areas of suspected elevated radionuclides also were sampled for this extended list of constituents. Results from this study were compared to drinking-water criteria as well as data from other large-scale USGS water-quality investigations.

As a consequence of the pervasive occurrence of uranium and other radioactive constituents in the natural environment, radioactive constituents were detected in each of the wells sampled during this study regardless of well location or geologic unit. Overall, nearly 41 percent (52 of the 128) of the sampled wells had at least one constituent that exceeded U.S. Environmental Protection Agency (USEPA) drinking-water standards or screening levels. Uranium concentrations were higher than the USEPA maximum contaminant level (MCL) of 30 µg/L in samples from 18 of 128 wells (14 percent). Radon concentrations exceeded a proposed MCL of 4,000 picocuries per liter (pCi/L) in 27 percent of the wells. About 40 percent (42 of 104 wells) of the wells had gross alpha-particle activities (72-hour count) at or greater than a screening level of 15 pCi/L. The adjusted gross alpha-particle activity (calculated activity after subtracting activity associated with uranium based on a 0.67 picocurie per microgram conversion factor) exceeded the USEPA MCL of 15 pCi/L in samples from 26 wells at the 72-hour count and 13 wells at the 30-day count. Gross beta-particle activity exceeded the 50 pCi/L screening level in 5 of 104 wells. Samples from 10 of 47 wells exceeded the 5 pCi/L combined radium (radium-226 and radium-228) MCL. U.S. Environmental Protection Agency currently (2013) does not have a MCL for polonium-210 and most of the 40 wells sampled had values less than 0.10 pCi/L.

The Boulder batholith is a known source of uranium-bearing minerals and is a common geologic unit found in the study area. Established or proposed drinking-water standards were exceeded most frequently in water from wells completed in the generalized geologic unit (TKi) that includes rocks of the Boulder batholith and other Tertiary through Cretaceous igneous intrusive rocks (commonly described as granite). Specifically, of the wells completed in the Boulder batholith and related rocks sampled as part of this study, 24 percent exceeded the MCL of 30 µg/L for uranium, 50 percent exceeded the proposed alternative MCL of 4,000 pCi/L for radon, and 27 percent exceeded the MCL of 5 µg/L for combined radium-226 and radium-228. Although median radionuclide concentrations were typically highest in samples from the Boulder batholith and similar units, elevated values were determined in other geologic units as well. Radiochemical constituents exceeded at least one current (2013) or proposed drinking-water standard or screening level in 5 of 6 generalized geologic units assessed during this study. Radiochemical-constituent concentrations were variable within all geologic units analyzed, including wells in close proximity to one another (less than 1,000 ft apart).

Elevated radionuclides occurred over nearly the entire range of physical properties and water types. Correlations between radioactive constituents and pH, dissolved oxygen, total dissolved solids, and major-ion chemistry were not statistically significant (p-value >0.05) or were considered only weakly to moderately correlated. Moderate correlations, although the strongest between radiochemical constituents and major ions, occurred between potassium and gross beta-particle activity (rho = 0.72 to 0.82) as gross beta-particle activities typically increased with increased potassium concentrations. Because potassium (specifically potassium-40) is a beta-particle emitter it is a likely contributor to the gross beta-particle activity in these waters. Total dissolved solids also was moderately correlated (rho = 0.63 to 0.71) with gross alpha-particle and gross beta-particle activity, indicating that higher radioactivity values can be associated with higher total dissolved solids. The strongest correlations among radioactive constituents occurred among the radium isotopes (rho = 0.78 to 0.92) and among the gross alpha-particle and beta-particle activities (rho = 0.77 to 0.96). Correlations of gross alpha-particle activity (72-hour count) were statistically significant (p-value <0.05) with all analyzed radioactive constituents and therefore gross alpha-particle activity may be considered as a screening tool to detect the presence of radionuclides in area groundwater. Gross alpha-particle activities (72-hour count) of 7 pCi/L or higher were associated with all radioactive constituents that exceeded drinking-water standards or screening levels in this study.

Data from this study illustrate the temporal variability of water chemistry in area wells and the potential effect of holding time duration on results, interpretation, and decisions regarding actions relative to water-quality criteria or concerns. Temporal variability of water quality was assessed by sampling a subset of wells on more than one occasion. Relative percent differences of greater than 30 percent (considered as large temporal variability for this report) for multiple analytes suggest the need for multiple samples collected over a range of conditions throughout time to determine representative water-quality conditions. Large temporal variation of gross alpha-particle activities were observed among 38 percent of the resampled wells. Other constituents that had large temporal variability among resampled wells include: uranium (29 percent of the resampled wells), gross-beta particle activity (15 percent of the resampled wells), and radon (11 percent of the resampled wells). These large temporal variations occurred at all sampling intervals, whether it was 1–2 months, 10 months, or 3 years.

Gross alpha- and beta-particle activities from this study illustrate that holding times before sample analysis can affect results, interpretation, and decisions regarding actions relative to water-quality criteria or concerns. Gross alpha- and beta-particle activities were counted 72 hours and again 30 days after sample collection. Gross alpha-particle activity decreased on average about 37 percent in samples measured after 72 hours and again after 30 days. The decrease in gross alpha-particle activity indicates the presence of short-lived alpha-emitting radionuclides in the sampled waters. Gross beta-particle activity increased on average about 31 percent in samples measured at 72 hours and again at 30 days. This

increase in gross beta-particle activity over that time period indicates the ingrowth (incremental accumulation of daughter products generated by the decay of the parent) of beta-emitting progeny of uranium.

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Glossary

Many of the definitions in the glossary are taken from Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP) (U.S. Environmental Protection Agency and others, 2004), McCurdy and others (2008), Szabo and others (2005), and USGS National Water-Quality Assessment Program Radium Glossary web page accessed October 2, 2012, at *http://water.usgs.gov/nawqa/trace/radium/glossary.html*.

activity   The amount of radioactivity in a given volume of such material as water or air. Mean rate of nuclear decay occurring in a given quantity of material. The "Curie" unit for activity is currently (2013) used by the USGS. The SI unit of activity is the becquerel (Bq), which equals one nuclear transformation per second. One curie equals 3.7×10^{10} Bq. 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 to contact 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 alpha-particle radioactivity emitted by isotopes of naturally occurring uranium, thorium, radium, and progeny such as polonium. Long-term measurement, usually conducted from 20 to 30 days after sample collection, measures only the amount of alpha radiation present from long-lived radionuclides.

alpha radioactivity count   An analytical technique that specifies alpha radioactivity emitted by a chemically purified sample that contains only one radionuclide of interest. The alpha-particle activity is counted in a low-background gas proportional counter. If the "chemically purified sample" contains polonium-210 as the only radionuclide of interest, it would be counted by alpha spectrometry, not gas proportional counter.

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).

background (instrument)   Radiation detected by an instrument when no source is present. The background radiation that is detected may come from radionuclides in the materials of construction of the detector, its housing, its electronics, and the building as well as the environment and natural radiation.

beta-particle radioactivity   Beta-particle 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-particle radiation, like alpha radiation, is ionizing radiation—it strips electrons from adjacent atoms as it passes. Beta-particle radiation can penetrate only the surface layer of skin; thus, a beta-particle emitting radionuclide must be ingested to contact internal organs or tissues. An accumulation of tissue damage in the cell nucleus may lead to cell mutation and potential cancer formation.

beta-particle radioactivity, gross   A laboratory measurement of total beta-particle radioactivity emitted by a sample. This measurement includes radioactivity emitted by naturally occurring progeny of uranium and thorium, such as radium-228 and lead-210, and numerous other naturally occurring beta-particle-emitting radionuclides such as potassium.

beta-particle radioactivity count An analytical technique that specifies total beta-particle 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.

blank (analytical or method) A sample that is assumed to be essentially free of the radionuclide that is carried through the radiochemical preparation, analysis, mounting, and measurement process in the same manner as a routine sample of a given matrix.

combined standard uncertainty (CSU) The standard uncertainty of an output estimate calculated by combining the standard uncertainties of the input estimates. The combined standard uncertainty of y is denoted by $u_c(y)$. The CSU is reported at the 68 percent or 1-sigma (1σ) confidence level.

critical level (LC) In the context of analyte detection, critical level means the minimum measured value (for example, of the instrument signal or the radionuclide concentration) that indicates a positive (nonzero) amount of a radionuclide is present in the material within a specified probable error. The critical level is sometimes called the critical value or decision level. The general use of the term "critical level" is for a method wherein nominal measurement parameters are used in the calculation. Contrast this use to the sample-specific critical level (ssLC) defined herein.

decay factor The fractional amount of the original radionuclide activity in a sample that remains after decay in the time interval between sample collection and sample analysis.

gamma radioactivity Gamma radiation is composed of packets of energy, also known as photons or photon particles, termed collectively as gamma rays, 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-particle radiation, which cannot. An accumulation of tissue damage in the cell nucleus from gamma radiation may lead to cell mutation and potential cancer formation.

half-life The interval of time required for exactly one-half the atoms of a given radionuclide to undergo radioactive decay.

ingrowth The incremental accumulation of daughter products generated by the decay of the parent through time.

ingrowth factor The activity of a supported radionuclide progeny at a specific time after chemical separation, expressed as a fraction of the amount of radioactivity at full ingrowth.

ionizing radiation Consists of particles or photons that have enough energy to ionize an atom or molecule by completely removing an electron from its orbit, thus creating a more positively charged atom.

isotope Any of two or more forms of a single element having the same number of protons in their nuclei (same atomic number), but differing in the number of neutrons (different mass numbers). Unstable isotopes of an element decay through the emission of a form of radioactivity.

laboratory reporting level (LRL) The laboratory reporting level is a minimum concentration pre-determined by the laboratory personnel before the sample analysis begins; generally, it is based on typical counting results under routine analytical instrument operating conditions in a given sample matrix using known standards and blank samples, which the laboratory personnel define as a level of detection they can routinely achieve.

maximum contaminant level (MCL) A regulatory limit established by the U.S. Environmental Protection Agency (USEPA) for the concentration of certain radionuclides in drinking water. The highest level (concentration) of a contaminant that is allowed in drinking water distributed to the public. MCLs are set as close as feasible to the level believed to cause no human health effects, while using the best available treatment technology and taking cost into consideration. MCLs are enforceable standards.

minimum detectable concentration (MDC) The minimum detectable concentration of the analyte in a sample. The smallest (true) radionuclide concentration that gives a specified probability (Type II -) that the value of the measured radionuclide will exceed its critical level concentration (that is, that the material analyzed is not "blank" or free of analyte) (U.S. Environmental Protection Agency and others, 2004, chapter 20). The general use of the term "MDC" or "a priori MDC" is for a method wherein nominal measurement parameters are used in the calcula-

tion. Contrast this use to the sample-specific MDC (ssMDC) defined herein.

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 long-term average background radiation reported routinely by the instrument. In practical terms, the result is less than the sample-specific minimum detectable concentration and indicates that radioactivity was not detected.

nuclide   A species of atom, characterized by its mass number, atomic number, and nuclear energy state, providing the mean half-life in that state is long enough to be observable.

picocurie per liter   The measurement unit that expresses the amount of radioactivity in water. One picocurie per liter (pCi/L) equals 2.2 radioactive disintegrations per minute per liter of water.

precision estimate (PE) or combined standard uncertainty (csu)   A calculated measure of uncertainty of the laboratory analysis. Determination of the precision estimate for radionuclides evaluates many sources of error, some that are unique to radionuclides. 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 the "precision estimate" or the "combined standard uncertainty (CSU)" (or less frequently as the "total propagated uncertainty"), and (2) a counting error or counting uncertainty only. There is a 67 percent or 95 percent probability (based on one or two standard deviations, respectively, of the radioactivity count) that the true value of the radionuclide concentration is within the range of the reported measured value plus or minus the precision estimate or uncertainty. The precision estimate or uncertainty terms generally are less than the measured value except when the measured values are low concentrations (near the LRL). The precision estimates associated with the various radionuclide concentrations determined by the alpha spectrometry measurement technique and presented in this report are given as the two standard deviation propagated precision estimates.

quality control (QC)   The overall system of technical activities that measures the attributes and performance of a process, item, or service against defined standards to verify that they meet the stated requirements established by the project; operational techniques and activities that are used to fulfill requirements for quality. This system of activities and checks is used to ensure that measurement systems are maintained within prescribed limits, providing protection against out-of-control conditions and ensuring that the results are of acceptable quality.

radioactivity   The property possessed by some elements or isotopes of spontaneously emitting energetic particles (electrons or alpha particles) by the disintegration of their atomic nuclei.

radiochemical constituent   An inclusive term used in this report to include radionuclides and emitted particles.

radiogenic   Resulting from radioactive decay, usually in reference to natural products such as lead.

radiochemistry   Branch of chemistry that deals with radioactive materials.

radiogenic daughter product   The product resulting from the radioactive disintegration or nuclear transformation of its parent radionuclide.

radiological hold time   Refers to the time differential between the sample collection date and the final sample counting (analysis) date.

radionuclide   A nuclide that is radioactive (capable of undergoing radioactive decay).

sample-specific critical level (ssLC)   The sample-specific critical level is calculated using the parameter values measured during the generation of the sample result. This is different than the critical level for a method wherein nominal measurement parameters are used in the calculation (see critical level). Concentrations less than the ssLC are considered nondetections.

sample-specific minimum detectable concentration (ssMDC)   The sample-specific minimum detectable concentration is calculated using the parameter values measured during the generation of the sample result (see minimum detectable concentration).

secular equilibrium   Radioactive equilibrium when the half-life of the progeny is much

less than the half-life of the parent. A situation in which the quantity of a radionuclide remains constant because its production rate (for example, the decay of a parent isotope) is equal to its decay rate.

standard uncertainty   The uncertainty of a measured value expressed as an estimated standard deviation, often called a "1-sigma" (1-σ) uncertainty. The standard uncertainty of a value x is denoted by u(x).

uncertainty   A parameter, usually associated with the result of a measurement, which characterizes the dispersion of the values that could reasonably be attributed to the measurement of interest.

Appendixes 1–8

Appendix 1. Site-Identification System

Groundwater sites (wells) are assigned a 15-digit site identification number; these numbers represent the approximate latitude and longitude of the site (first 13 digits) plus a sequence number (last 2 digits). Wells used for this study also were assigned a station name based on their geographic position within the rectangular grid system used for the subdivision of public lands (fig. 1–1). The station name consists of 14 characters. The first three characters specify the township and its position north (N) of the baseline in Montana (for example, 09N). The next three characters specify the range and its position east (E) of the principal meridian in Montana (for example, 05E). The next two numbers represent the section number. The next four characters sequentially designate the quarter (160-acre tract), quarter-quarter (40-acre tract), quarter-quarter-quarter (10-acre tract), and the quarter-quarter-quarter-quarter (2 ½-acre tract) of the section in which the well lies. The quarter subdivisions within a section are designated A, B, C, and D in a counterclockwise direction, beginning in the northeast quadrant. The final two characters of the station name are a sequence number assigned to differentiate multiple wells within a single quarter-quarter-quarter-quarter section. For example, as shown in figure 1–1, 09N05E18BBAC01 was the first well inventoried in the SW1/4 of the NE1/4 of the NW1/4 of the NW1/4 of the SE1/4 of sec. 18, T. 09N., R. 05E.
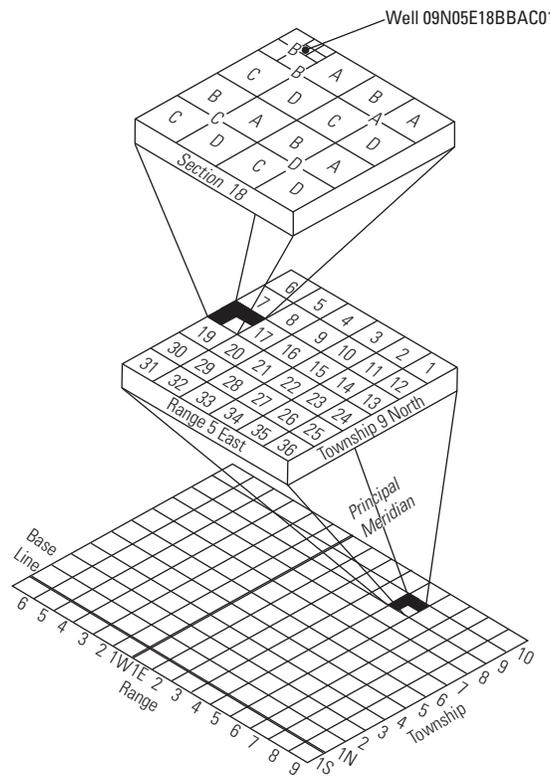

Figure 1–1.   Site-identification system for wells.

Appendixes 2–8

Appendixes 2–8 are tables in an Excel spreadsheet. The Excel file is available at *http://pubs.usgs.gov/sir/2013/5235.*

Appendix 2.   Records of wells sampled for the study of radiochemical constituents in groundwater of Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 3.   Physical properties and inorganic chemistry of groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 4.   Analyses of inorganic chemistry of field replicates of groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 5.   Analyses of physical properties and inorganic chemistry of field-blank samples associated with groundwater sampling from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 6.   Radiochemistry of groundwater samples collected from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 7.   Radiochemistry of field-replicate samples and associated environmental groundwater samples from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Appendix 8.   Radiochemistry of field blank samples associated with groundwater sampling from Jefferson County and surrounding areas, Montana, 2007 through 2010.

Back cover photographs.   Top, domestic well completed in the Boulder batholith and frost-free hydrant (preferred sampling point) near Clancy, Montana, January 20, 2014. Photograph by Heath R. Caldwell. Middle, groundwater sample collection in a mobile laboratory, September 27, 2010. Photograph by Rodney R. Caldwell, U.S. Geological Survey. Bottom, collection of groundwater sample for radon analysis, September 27, 2010. Photograph by Hannah L. Nilges, U.S. Geological Survey.

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