

Abstract

A study of uranium in groundwater in northeastern Washington was conducted to make a preliminary assessment of naturally occurring uranium in groundwater relying on existing information and limited reconnaissance sampling. Naturally occurring uranium is associated with granitic and metasedimentary rocks, as well as younger sedimentary deposits, that occur in this region. The occurrence and distribution of uranium in groundwater is poorly understood. U.S. Environmental Protection Agency (EPA) regulates uranium in Group A community water systems at a maximum contaminant level (MCL) of 30 µg/L in order to reduce uranium exposure, protect from toxic kidney effects of uranium, and reduce the risk of cancer. However, most existing private wells in the study area, generally for single family use, have not been sampled for uranium. This document presents available uranium concentration data from throughout a multi-county region, identifies data gaps, and suggests further study aimed at understanding the occurrence of uranium in groundwater.

The study encompasses about 13,000 square miles (mi<sup>2</sup>) in the northeastern part of Washington with a 2010 population of about 563,000. Other than the City of Spokane, most of the study area is rural with small towns interspersed throughout the region. The study area also includes three Indian Reservations with small towns and scattered population. The area has a history of uranium exploration and mining, with two inactive uranium mines on the Spokane Indian Reservation and one smaller inactive mine on the Okanogan Reservation.

Historical (1977–2016) uranium in groundwater concentration data were used to describe and illustrate the general occurrence and distribution of uranium in groundwater, as well as to identify data deficiencies. Uranium concentrations were detected at greater than 1 microgram per liter (µg/L) in 60 percent of the 2,382 historical samples (from wells and springs). Uranium concentrations ranged from less than 1 to 88,000 µg/L, and the median concentration of uranium in groundwater for all sites was 1.4 µg/L.

New (2017) uranium in groundwater concentration data were obtained by sampling 13 private domestic wells for uranium in areas without recent (2006) water-quality data. Uranium was detected in all 13 wells sampled for this study; concentrations ranged from 1.03 to 1,180 µg/L with a median of 22 µg/L. Uranium concentrations of groundwater samples from 6 of the 13 wells exceeded the MCL for uranium. Uranium concentrations in water samples from two wells were 1.10 and 1,180 µg/L, respectively; nearly 40 times the MCL.

Additional data collection and analysis are needed in rural areas where self-supplied groundwater withdrawals are the primary source of water for human consumption. Of the roughly 43,000 existing water wells in the study area, only 1,255 wells, as summarized in this document, have available uranium concentration data, and some of those data are decades old. Furthermore, analysis of area groundwater quality would benefit from a more extensive chemical-analysis suite including general chemistry in order to better understand local geochemical conditions that largely affect the mobility of uranium. Although the focus of the present study is uranium, it also is important to recognize that there are other radionuclides of concern that may be present in area groundwater.

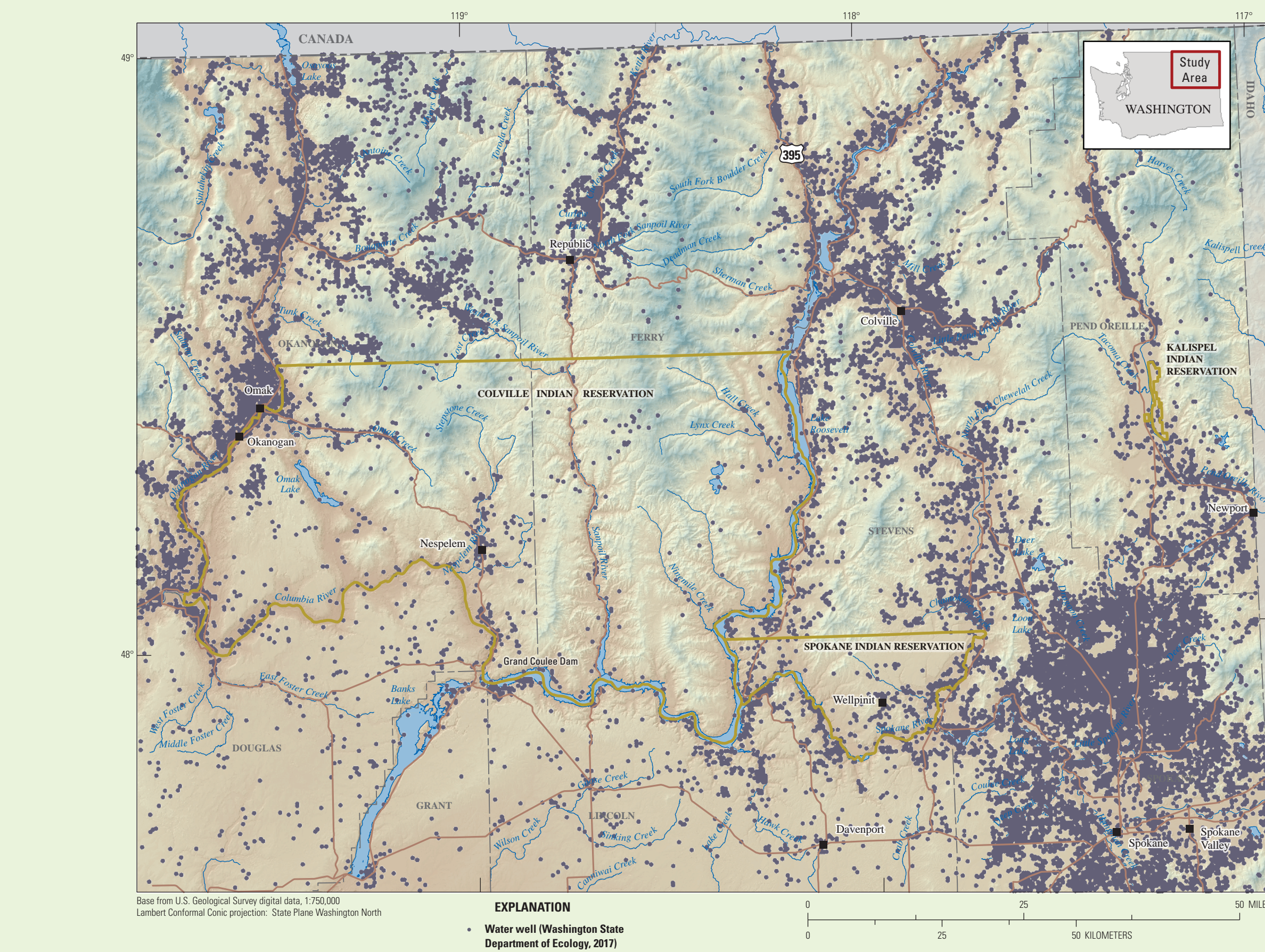


Figure 1. Location of study area and areal distribution of existing water wells included in the Washington State Department of Ecology well-log database (Washington State Department of Ecology, 2017), northeastern Washington.

Uranium in the Environment

Uranium is a radioactive element, or radionuclide, that occurs naturally, generally in low concentrations, in rock, soil, surface water and groundwater (Argonne National Laboratory, 2007). Unlike many water pollutants, all radionuclides dissolved in water are colorless, odorless, and cannot be detected by our senses (Zapeca, and Szabo, 1988). Uranium can be highly concentrated in intrusive igneous rocks, primarily granitic rocks, and also is found in sedimentary rocks such as black shales, silt to silts, and phosphates (Ayyotte and others, 2007, 2011).

Uranium-238 is the most common of several natural isotopes and accounts for 99 percent of the mass of natural samples. Uranium decays slowly by emitting alpha particles and has multiple daughter products and decay paths (fig. 2). Radiation, in the form of alpha and beta particles, cannot be detected by our senses. The most stable nuclei of uranium and its daughter products. Figure 2 shows the important daughter products in the U-238 decay series, indicates whether the primary decay mode is through alpha or beta emission, and gives the half-life (length of time required for half of the atoms of a radioactive isotope to decay). For example, through the decay series of U-238, alpha and beta particles are emitted and radium and radon are formed as interim radioactive daughter products. These, in turn, ultimately decay to a stable form of lead.

Federal Regulation

EPA's Final Rule for Radionuclides in Drinking Water took effect in 2003, regulating uranium at a maximum contaminant level (MCL) of 30 µg/L (U.S. Environmental Protection Agency, 2000). The rule also regulates radium, and alpha and beta particles due to their radiological toxicity. Group A community water systems, which serve at least 15 service connections or 25 residents regular year around, are required to meet the final MCLs in order to reduce uranium exposure, protect from toxic kidney effects of uranium, and reduce the risk of cancer (U.S. Environmental Protection Agency, 2001).

Geologic Setting and Uranium Exploration

The geology of northeastern Washington is complex and is composed of various types of bedrock (for example, shale, slate, dolomite, quartzite, and granite) overlain in many places with varying types and thicknesses of unconsolidated sediment such as silt, sand, gravel, and clay (fig. 3). Bedrock units in the area record a complex structural history involving repeated periods of folding, faulting, and igneous intrusion (Lamont, 1991).

Uranium ore was discovered in Washington in the 1950s, and was mined at several locations including the Midvale and Sherwood Mines on the Spokane Reservation in Stevens County, and the smaller Daybreak Mine on the western side of Mount Spokane in Spokane County (fig. 3). Uranium assays (prospects) also occur throughout the region (fig. 3). Extraction of uranium ore at these mines largely ended by the early 1980s (Dahlkamp, 2011).

The uranium ore at the Midvale Mine was located in a contact zone between a granitic intrusion and surrounding metasedimentary rocks. At the Sherwood Mine, the uranium ore was located in conglomerate overlying granite where the mineralization was associated with carbonaceous matter within the conglomerate matrix (Dahlkamp, 2011). At the Daybreak Mine, uranium minerals were concentrated along shear zones and within pegmatite dikes within granitic rocks, including medium-quality autinite (hydrated calcium uranyl phosphate) crystals (Dahlkamp, 2011). Weathering of uranium-bearing units such as the granites resulted in occurrence of uranium in younger sedimentary deposits, including a small post bog near Colville (Johnson and others, 1987). Due to the presence of uranium-rich granitic rocks, as well as the general mobility of uranium in circulating groundwater, the regions that are likely susceptible to occurrences of uranium in groundwater include Pend Oreille, Stevens, and Ferry Counties, as well as eastern Okanogan, northern Spokane, and northeastern Lincoln Counties (fig. 3).

Historical Data Sources

Although much of the study area is remote and sparsely populated, the bedrock aquifers in northeastern Washington are increasingly being developed for drinking-water supply. In most situations, private wells are only tested at the well owner's discretion or when they are newly constructed. In order to inform owners of private and unregulated wells, two fact sheets were developed to encourage private well owners to test the water from their wells for uranium and their indoor air for radon gas (Northeast Tri-County Health District and Washington State Department of Health, 2006; Washington State Department of Health, 2011). Historical (1977–2016) uranium concentration data were obtained from various local and national sources to describe and illustrate the general occurrence and distribution of uranium in groundwater in northeastern Washington.

Local Sources

The presence of naturally occurring uranium in local groundwater became evident in the early 2000s following selected testing of new or existing water sources, many near Colville in Stevens County, Washington (Northeast Tri-County Health District (NETCHD) and Washington State Department of Health (WDOH), 2006; Schneider, 2016). Due to increasing concerns about the possible presence of uranium in area groundwater, the NETCHD added uranium to their water-quality testing recommendations in 2012 in the three counties it serves—Ferry, Stevens, and Pend Oreille (M. Schanz, NETCHD, written commun., 2013). By November 2016, samples from 411 private wells had been collected in the Tri-County area. In 2017, the NETCHD Board of Health added initial testing of uranium for Group B public water-supply systems (which generally serve 3 to 14 connections or fewer than 25 people per day) in

Introduction

Groundwater monitoring in northeastern Washington has shown elevated concentrations of uranium in private wells and several community water systems. Naturally occurring uranium is associated with granitic and metasedimentary rocks, as well as younger sedimentary deposits, that occur in this region. The occurrence and distribution of uranium in groundwater is poorly understood and warrants investigation in order to reduce human exposure to uranium, protect kidneys from toxic effects of uranium, and reduce the risk of cancer. Basic knowledge and a greater awareness of the occurrence and distribution of uranium in groundwater throughout northeastern Washington would aid homeowners and health officials in evaluating its potential presence and making informed decisions on further testing and treatment of drinking water supplies.

Purpose and Scope

This document presents available uranium concentration data from throughout a multi-county region, identifies data gaps, and suggests further study aimed at understanding the occurrence of uranium in groundwater. It also conveys the issue and presents available information to local health agencies and the general public in order to reduce exposure of humans to potentially harmful naturally occurring uranium in groundwater. Historical geologic, hydrogeologic, and uranium concentration data for groundwater for an area of northeastern Washington was compiled from multiple sources, tabulated, and mapped. Recent (2017) groundwater samples were collected by the U.S. Geological Survey (USGS) and analyzed for uranium concentrations from 13 wells in areas without recent water-quality data. The distribution of uranium concentrations in groundwater was evaluated with regard to the geology; the material within the water-bearing zone of the sampled water wells and the material mapped at land surface at the well and spring locations.

Description of Study Area

The study area includes all of Ferry, Stevens, and Pend Oreille Counties, most of Spokane County, and parts of Okanogan, Douglas, Grant, and Lincoln Counties, and encompasses about 13,000 mi<sup>2</sup> in the northeastern part of Washington (fig. 1). Based on the 2010 Census, the population of the study area is about 563,000, the largest city is Spokane, Washington, with a population of about 209,000 (Washington State Office of Financial Management, 2017). With the exception of Spokane, most of the study area is rural with small towns interspersed throughout the region. A population of about 438,000 in the study area is served by public water systems, and 22 percent, or a population of about 125,000, is served by private domestic wells (Washington State Department of Health, 2017; Washington State Office of Financial Management, 2017). The distribution of the roughly 43,000 existing water wells in the study area is shown in figure 1.

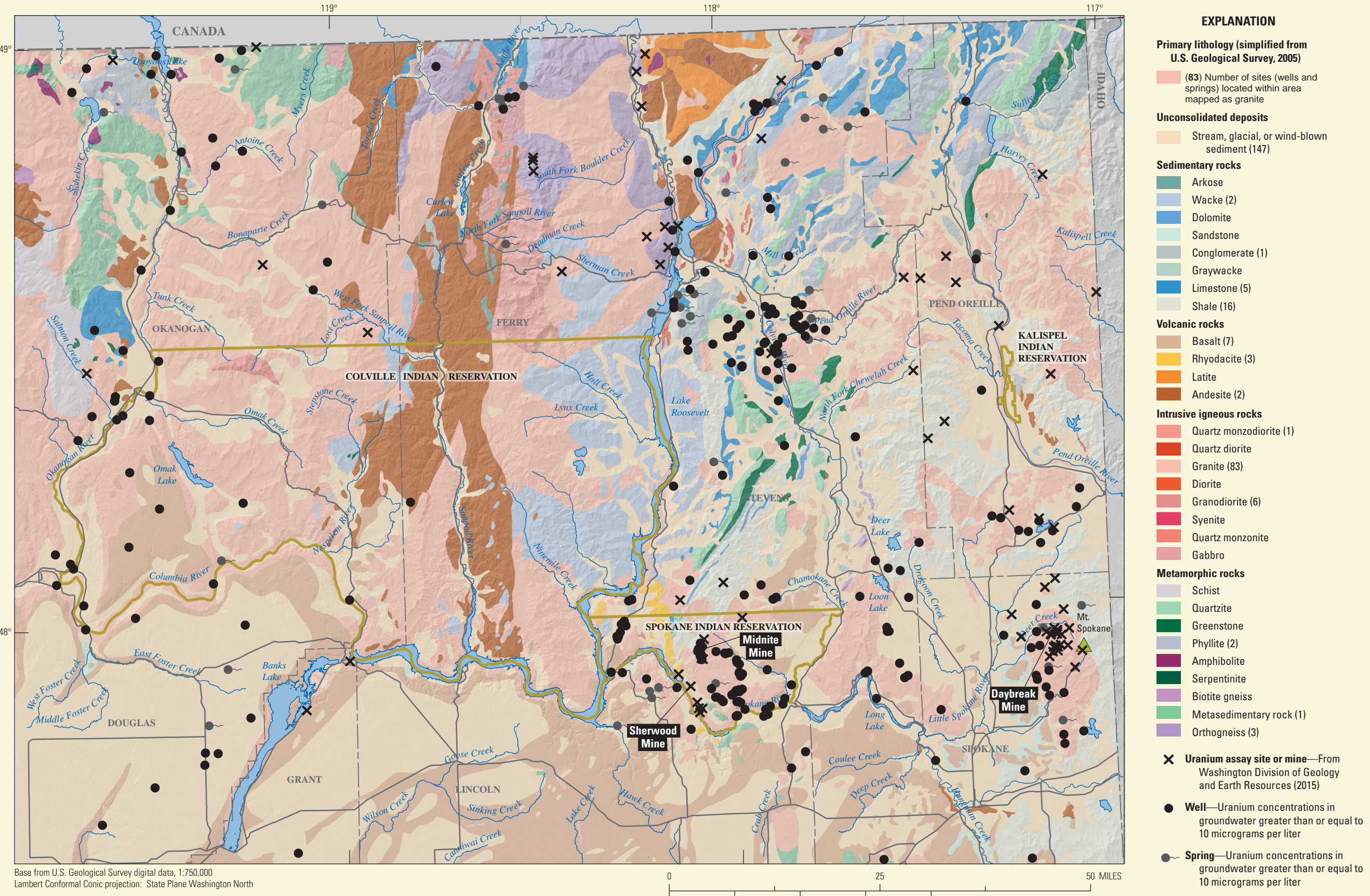


Figure 3. Geology, locations of uranium assay sites or mines, and locations of wells and springs with historical uranium concentrations in groundwater of greater than or equal to 10 micrograms per liter (µg/L), northeastern Washington, 1977–2016.

Uranium Concentrations in Groundwater

Historical (1977–2016) uranium concentration data were compiled from NETCHD, the Spokane Tribe, USGS (NWIS and NURE), and WDOH and were used to describe and illustrate the general occurrence and distribution of uranium in groundwater in northeastern Washington. The data are from a total of 2,382 sites (wells and springs) and are listed in table 1, summarized in table 2, and shown in figure 4 by ranges of uranium concentrations. Uranium concentrations were detected at greater than 1 µg/L in 60 percent of the 2,382 historical samples. Uranium concentrations ranged from less than 1 to 88,000 µg/L, and the median concentration of uranium in groundwater for all sites was 1.4 µg/L. A total of 84 sites sampled (4 percent) exceeded EPA's MCL of 30 µg/L uranium (table 2, fig. 4). Uranium concentrations ranged from less than 1 to 965 µg/L, not including the results for the monitoring wells near Midvale Mine.

Most historical data were collected during the late 1970s as part of the NURE program to evaluate uranium resource potential rather than to assess groundwater quality. The NURE dataset does, however, provide the most widespread coverage of historical concentration data for uranium in groundwater. The remaining 763 historical sites were sampled specifically for water-quality purposes, and those data were from WDOH, USGS, NETCHD, and the Spokane Tribe (table 2). Stevens County, with data for 563 wells, had the greatest coverage compared to all counties in northeastern Washington due to the adoption of testing in 2012 through their building permit process, as well as the Spokane Tribe's data collection that began at the same time.

Table 2. Summary of historical uranium concentrations in groundwater including number of sites in each dataset, range of sample collection dates, and range of values, northeastern Washington, 1977–2016.

(Source: data from Washington State Department of Health, NURE, National Uranium Resource Evaluation program; NWIS, U.S. Geological Survey National Water Information System; NETCHD, Northeast Tri-County Health District; Spokane Tribe, Spokane Tribe of Indians. Uranium concentration: Reported in micrograms per liter (µg/L). Minimum value, median value, maximum value, (see text). Number of sites  $\geq 30$  µg/L. Number of sites with uranium concentrations greater than or equal to 30 micrograms per liter.)

Source of data	Area sampled (acres)	Number of sites	Range of sample collection dates	Uranium concentration (µg/L)	Number of sites $\geq 30$ µg/L (percent)	Notes
DOH	All of study area	47	1994–2016	[1; 8.25; 49.6]	8 (17)	Most recent untreated unbanded sample when available (3 samples from well fields when samples from individual wells not available).
NURE (USGS)	All of study area	980	1977–1979	[1; 17.1; 165]	22 (1)	One-time sample during national uranium exploration.
NWIS (USGS)	All of study area	67	1980–2016	[1; 2.18; 88,600]	15 (22)	Most recent sample; sites are a mix of monitoring, domestic, and public supply wells. Highest values are associated with monitoring wells near Midvale Mine.
NETCHD	Ferry Pend Oreille Stevens	12 73 326	2012–2016 2012–2016 2008–2016	[1; 2.5; 965]	28 (7)	Owner-collected sample as part of building permit requirement or recommendation of Health Department (331 wells have driller's log).
Spokane Tribe	Spokane Reservation, Stevens	237	2012–2016	[1; 1.99; 203]	11 (5)	Initial sampling of all wells on the Reservation.
All sources	All of study area	2,382	1977–2016	[1; 1.4; 88,600]	84 (4)	Compilation of all data sources.

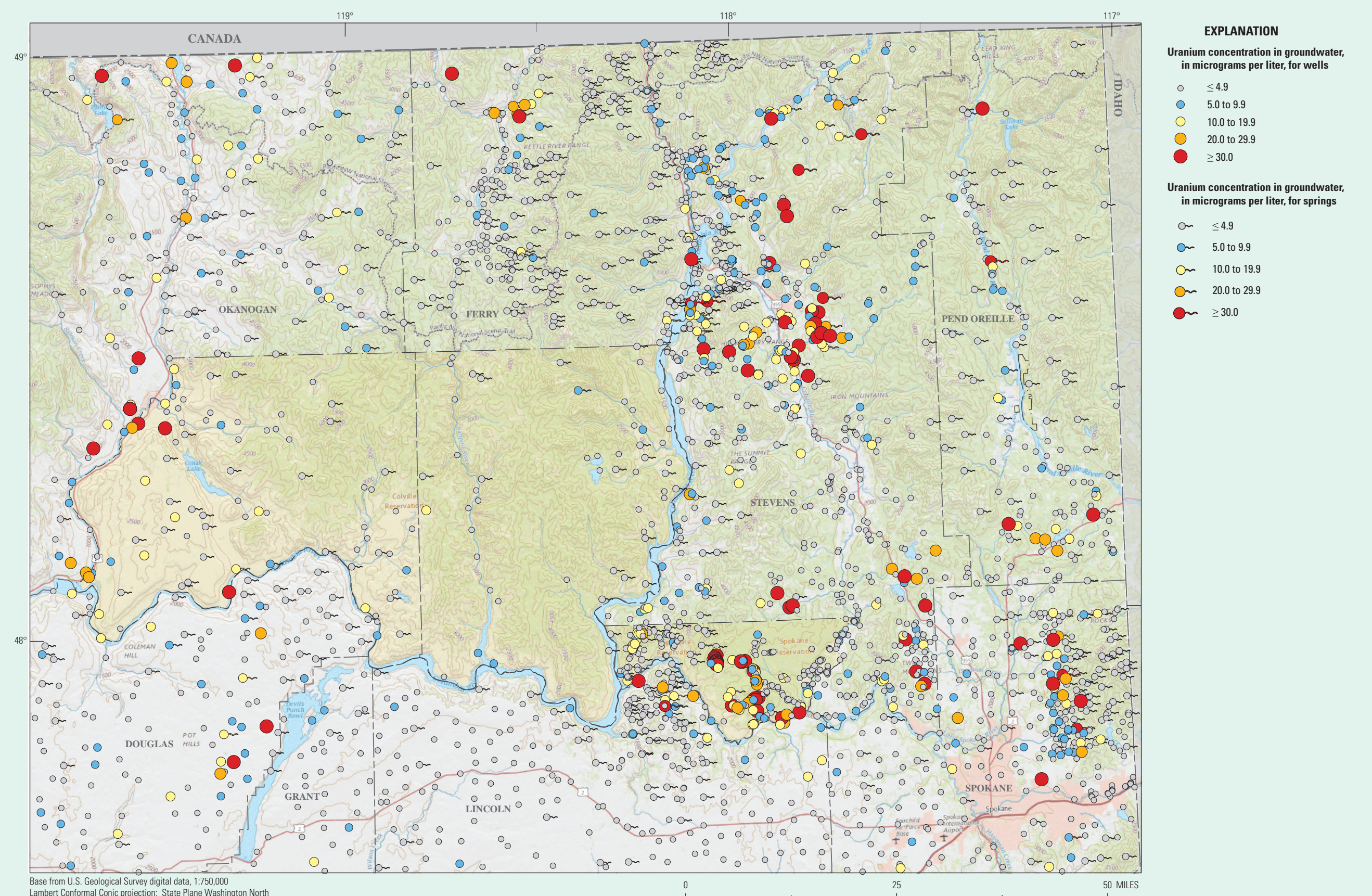


Figure 4. Magnitude and distribution of historical uranium concentrations in groundwater samples, northeastern Washington, 1977–2016.

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Comparison of Historical Uranium Concentrations with Geology

A surficial geologic map (fig. 3; U.S. Geological Survey, 2005) was used to examine if an association exists between the surficial geology mapped at sample sites and elevated concentrations of uranium (greater than or equal to 10 µg/L) in the water samples collected at those sites. There is a general, but inconsistent, relation between the locations of the 352 historical sample sites (wells and springs) with elevated uranium concentrations and mapped surficial geologic units (fig. 5). The relation exists for 83 sites that plot where granite is mapped and for 16 sites that plot where shale is mapped. However, the relation does not exist for 147 additional sites with elevated uranium that are located in the unconsolidated deposits mapped at the surface. In the areas where the sample sites are wells, these wells likely are open to and obtaining groundwater from deeper units, such as granite, than what is mapped at the land surface.

A better indicator for the potential occurrence of elevated uranium in groundwater produced from a well is the geologic material that supplies groundwater to the well. The lithologic description of the material that the wells were completed in (produced water from) was determined from the material described within the open intervals of the wells (locations of wells screens, perforations, or open bore holes) on the available driller's logs for 332 NETCHD wells. Lithologic descriptions on each log were simplified into the major rock type described within the open interval of the well. These generalized lithologic descriptions (example, granite) were then compared to the uranium concentrations of the groundwater samples collected at those wells. Summary statistics are provided in table 2 and locations of wells and well type (geologic material of open interval) are shown in figure 5. Uranium concentrations did not exceed the MCL (table 3) for wells completed in basalt (BT), limestone (LM), or quartzite (QT). However, uranium concentrations exceeded the MCL in 18 and 14 percent of wells completed in granite (GR) and shale (SH), respectively. Similarly, uranium concentrations exceeded the MCL in 12 percent of wells completed in multiple geologic units (M), but this was due to the wells being partially open to granite or shale. Uranium concentration was greater than the MCL in 1 of 130 wells completed in unconsolidated sediment, such as sand and gravel (SG).

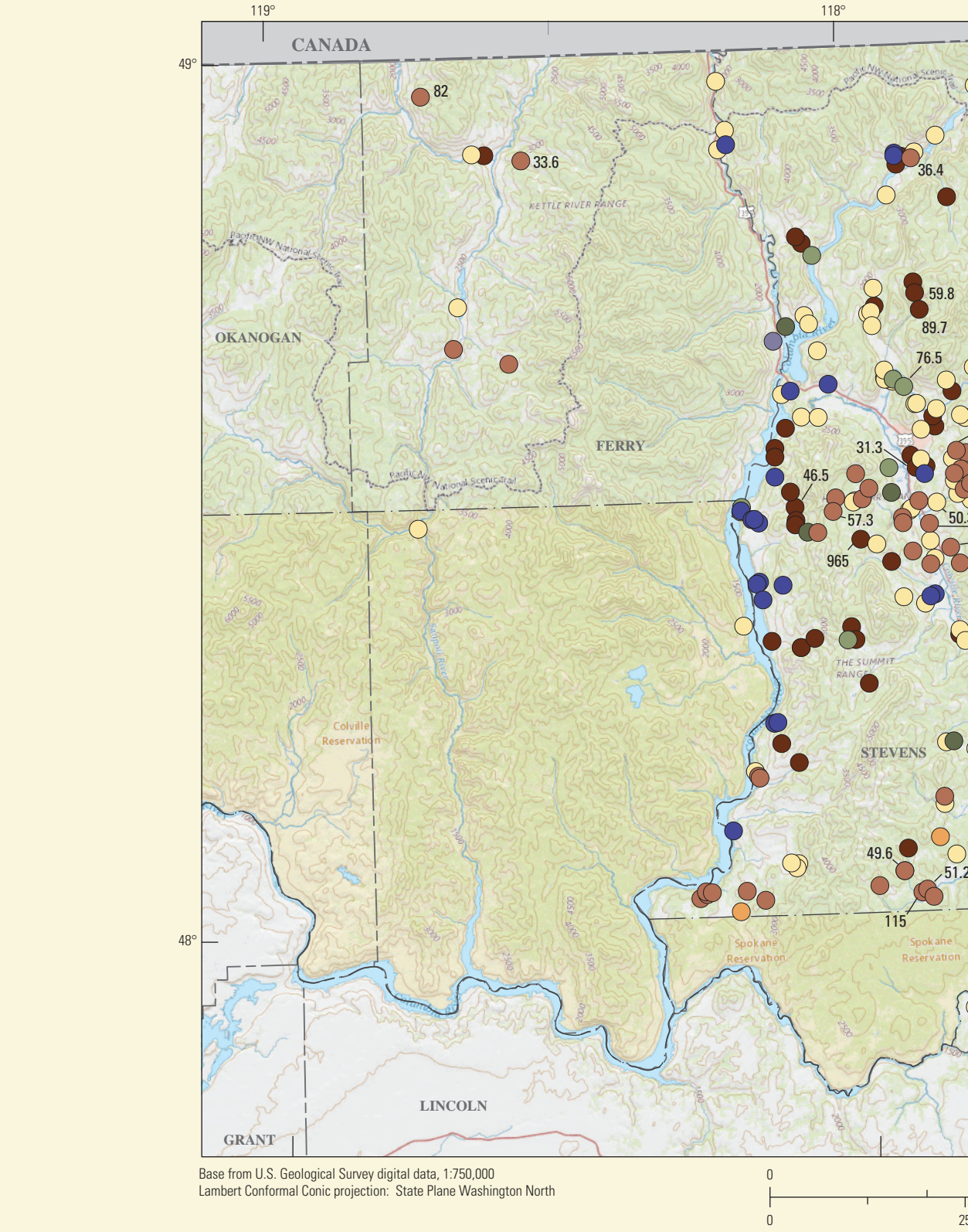


Figure 5. Locations of wells with associated uranium concentrations showing generalized geologic material of open interval, Ferry, Pend Oreille, and Stevens Counties, Washington. Wells with groundwater samples with uranium concentrations greater than or equal to 30 micrograms per liter are labeled.

Recent (2017) Uranium Concentration Data

The USGS collected groundwater samples from 13 private domestic wells for uranium analysis in July 2017 in areas without recent (2006) water-quality data but within areas where granite or shale was the predominant geology, mostly in northern Lincoln and Spokane Counties (fig. 6, table 4). The new data collected as part of this study was done in accordance with the USGS Washington Water Science Center water-quality assurance plan (Conn and others, 2017). Groundwater samples from the selected wells and associated quality-assurance samples were analyzed for uranium at the USGS National Water Quality Laboratory in Denver, Colorado, following inductively coupled plasma-mass spectrometry techniques described in Garbarino and others (2005). All data associated with these sample sites are stored in the USGS NWIS; site identifiers and results are included in table 4.

Uranium was detected in all 13 wells sampled for this study; concentrations ranged from 1.03 to 1,180 µg/L (tables 1 and 4) with a median of 22 µg/L. Uranium concentrations of groundwater samples from 6 of the 13 wells exceeded the MCL for uranium. Uranium concentrations in water samples from two wells (USGS02 and USGS11) were 1.10 and 1,180 µg/L, respectively; nearly 40 times the MCL.

The geographic distribution of the wells sampled shows by ranges of uranium concentrations are shown in figure 6. Only one (USGS07) of the three wells sampled in northern Lincoln County exceeded the MCL for uranium (table 4) and neither of the wells (USGS12 and USGS13) sampled in Stevens and Pend Oreille Counties exceeded the MCL for uranium (table 4). In northern Spokane County, however, five (USGS02, USGS04, USGS06, USGS09, and USGS11) of the eight wells sampled greatly exceeded the MCL for uranium with concentrations ranging from 152 to 1,180 µg/L.

Table 4. Uranium concentrations measured in groundwater samples with well depth and type, northeastern Washington, July 2017.

PROJ ID	USGS site identifier	Uranium concentration (µg/L)	Geologic material of open interval	Well depth (ft)	County
USGS01	4742848117191301	1.38	Granite	320	Spokane
USGS02	474607117114601	1,130	Granite	690	Spokane
USGS03	47471911707071	9.97	Granite	483	Spokane
USGS04	4749061171070501	152	Granite	473	Spokane
USGS05	474900117254101	5.3	Sand and gravel	64	Spokane
USGS06	47495311725501	160	Granite	517	Spokane
USGS07	475234118434401	18	Granite	498	Lincoln
USGS08	475253118495101	10.2	Granite	407	Lincoln
USGS09	47531211723901	218	Granite	298	Spokane
USGS10	47532511810401	10.6	Granite	598	Lincoln
USGS11	475545117135201	1,180	Granite	550	Spokane
USGS12	483422117485201	22	Shale	500	Stevens
USGS13	48522411725001	1.03	Shale	400	Pend Oreille

Considerations for Future Data-Collection Efforts

During the course of this investigation, several data needs were identified that, if filled, would provide a more complete understanding of the occurrence and distribution of uranium and other radionuclides of concern in northeastern Washington. Completion of these tasks could aid homeowners and health officials in evaluating the potential presence of harmful constituents and making informed decisions on further testing and treatment of drinking-water supplies. Potentially beneficial data-collection efforts identified during this study are listed below, in no particular order of importance.

- Much of the study area is under-represented in terms of available uranium concentration data for drinking-water sources. Additional data collection and analysis are needed in rural areas where self-supplied groundwater withdrawals are the primary source of water for human consumption. Most existing private wells in the study area have not been sampled for uranium. Of the roughly 43,000 existing water wells in the study area (fig. 1), only 1,255 wells, as summarized in this document, have available uranium concentration data, and some of those data are decades old.
- Although the focus of the present study is uranium, it is important to recognize that there are other radionuclides of concern that may be present in area groundwater. Based on limited data from regulated public supply systems and the Spokane Tribe, uranium concentrations in some wells were less than the standard, but radium and alpha particles were detected in some wells. Further testing of radionuclides in groundwater would provide area residents and local health agencies a

Table 3. Summary of historical uranium concentrations in groundwater samples by geologic material of open interval for 322 wells with available driller's logs, northeastern Washington.

Geologic material of open interval (unit abbreviation)	Median depth of well (ft)	Number of sites $\geq 30$ µg/L (percent)	Total number of sites	Uranium concentration (µg/L)
Basalt and unconsolidated interbeds (BT)	154	0 (0)	15	[1; <1; <2.50]
Granite (GN)	240	0 (0)	1	[16]
Granite (GR)	300	17 (18)	96	[<1; 4.68; 119]
Limestone (LM)	300	0 (0)	26	[<1; 1.86; 26.0]
Limestone (LS)	116	1 (1)	8	[1.11; 11.3; 36.5]
Quartzite or quartzite (QT)	300	0 (0)	9	[<1; 2.57; 9.03]
Muddy sand and gravel; some silt and clay (SG)	98	1 (1)	131	[<1; 2.44; 50.3]
Shale (SH)	280	6 (14)	43	[<1; 3.79; 965]

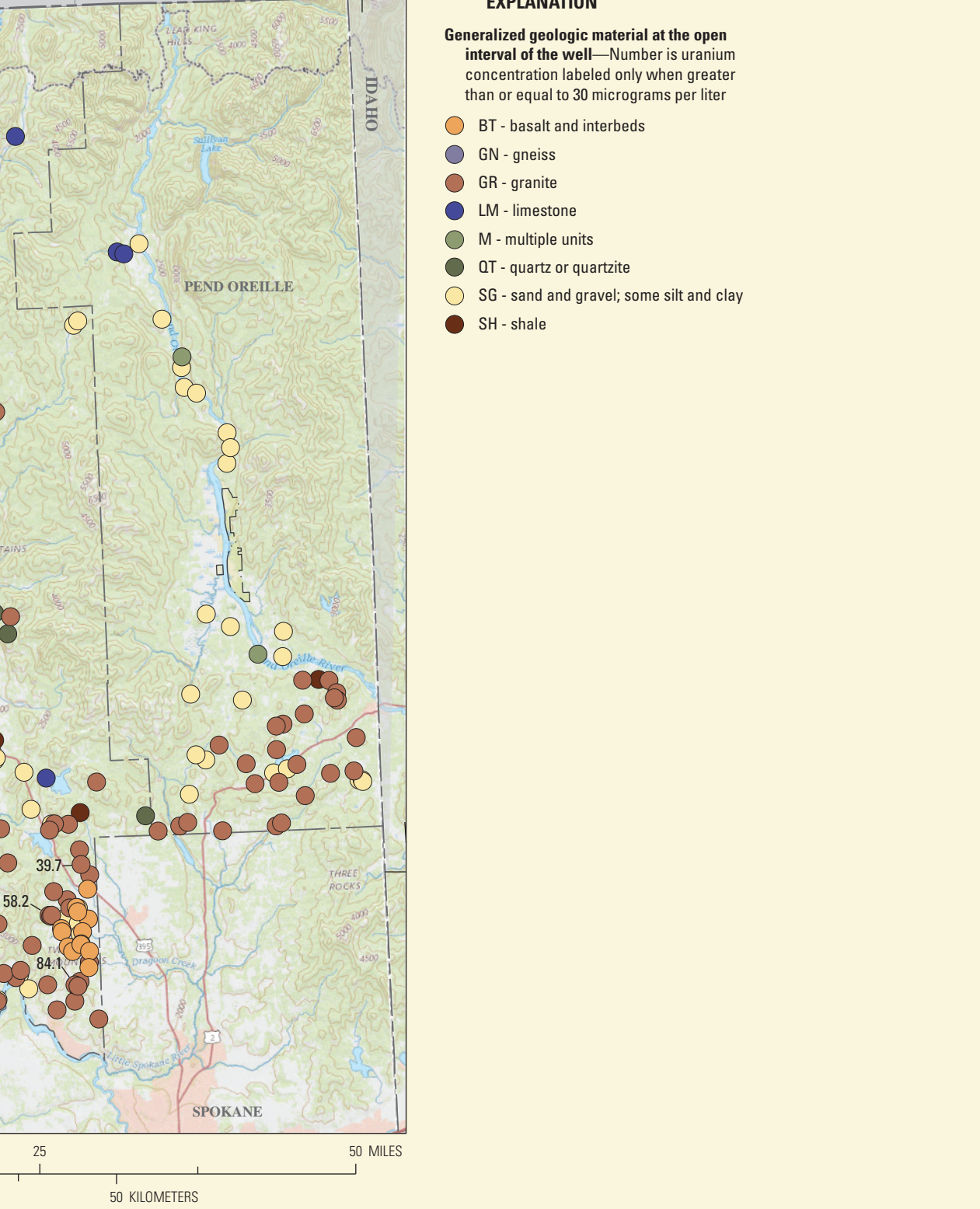
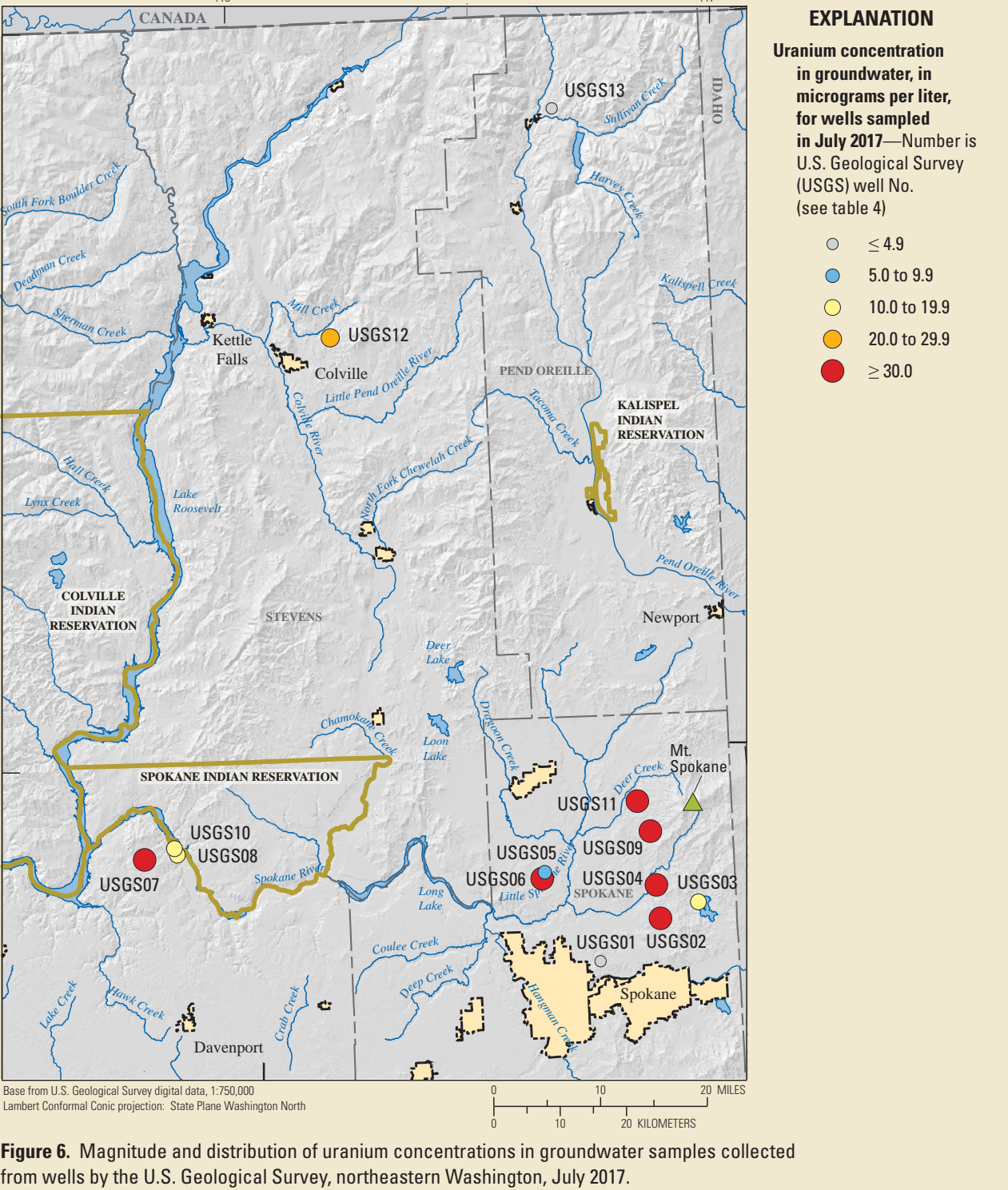


Figure 6. Magnitude and distribution of uranium concentrations in groundwater samples collected from wells by the U.S. Geological Survey, northeastern Washington, July 2017.



more complete understanding of their potential presence. Further, with sufficient data, screening protocols also might be developed to help address which wells, based on location, geology, and possibly depth, should be sampled and for which constituents.

- Further analysis of area groundwater quality would benefit from a more extensive chemical-analysis suite including general chemistry—pH, dissolved oxygen, common ion and organic carbon concentrations, and redox time—water quality. Further testing of radionuclides in groundwater would provide area residents and local health agencies a more complete understanding of their potential presence. Further, with sufficient data, screening protocols also might be developed to help address which wells, based on location, geology, and possibly depth, should be sampled and for which constituents.
- The occurrence of radionuclides in drinking water is not limited to Washington State but is rather a regional issue due to the occurrence of similar rock types in neighboring Idaho, Montana, and Canada. Recent studies in northern Idaho and western Montana document the presence of radionuclides at concentrations greater than health standards in drinking water (Agency for Toxic Substances and Disease Registry, 2006; Caldwell and others, 2013). A region-wide assessment utilizing available data would help raise awareness about the possible presence of radionuclides in the region's drinking water.

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