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Geochemical Analyses of Rock, Sediment, and Water from the Region In and Around the Tuba City Landfill, Tuba City, Arizona

By Raymond H. Johnson and Laurie Wirt

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Abbreviations

ppm = parts per million
ppb= parts per billion
µS/cm = microsiemens per centimeter
mg/L = milligrams per liter
µg/L = micrograms per liter

Conversion Factors

Inch/Pound to SI

Multiply	By	To obtain
	Length	
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)

SI to Inch/Pound

Multiply	By	To obtain
	Length	
meter (m)	3.281	foot (ft)
millimeter (mm)	0.03937	mile (mi)
	Volume	
liter (L)	33.82	ounce, fluid (fl. oz)
liter (L)	2.113	pint (pt)
liter (L)	1.057	quart (qt)
liter (L)	0.2642	gallon (gal)
liter (L)	61.02	cubic inch (in^3)
	Mass	
gram (g)	0.03527	ounce, avoirdupois (oz)

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

Horizontal coordinate information is referenced to the North American Datum of 1927 (NAD 27) in the UTM Zone 12N.

Specific conductance is given in microsiemens per centimeter at 25 degrees Celsius (µS/cm at 25°C).

Concentrations of chemical constituents in water are given either in milligrams per liter (mg/L) or micrograms per liter (µg/L).

Geochemical Analyses of Rock, Sediment, and Water from the Region In and Around the Tuba City Landfill, Arizona

By Raymond H. Johnson and Laurie Wirt

Abstract

The Tuba City Landfill (TCL) started as an unregulated waste disposal site in the 1940s and was administratively closed in 1997. Since the TCL closure, radionuclides have been detected in the shallow ground water. In 2006, the Bureau of Indian Affairs (BIA) contracted with the U.S. Geological Survey (USGS) to better understand the source of radionuclides in the ground water at the TCL compared to the surrounding region. This report summarizes those data and presents interpretations that focus on the geochemistry in the rocks and water from the Tuba City region.

The TCL is sited on Navajo Sandstone above the contact with the Kayenta Formation. These formations are not rich in uranium but generally are below average crustal abundance values for uranium. Uranium ores in the area were mined nearby in the Chinle Formation and processed at the Rare Metals mill (RMM). Regional samples of rock, sediment, leachates, and water were collected in and around the TCL site and analyzed for major and minor elements, ^{18}O, ^{2}H, ^{3}H, ^{13}C, ^{14}C, ^{34}S, ^{87}Sr, and $^{234}U/^{238}U$, as appropriate. Results of whole rock and sediment samples, along with leachates, suggest the Chinle Formation is a major source of uranium and other trace elements in the area. Regional water samples indicate that some of the wells within the TCL site have geochemical signatures that are different from the regional springs and surface water. The geochemistry from these TCL wells is most similar to leachates from the Chinle Formation rocks and sediments. Isotope samples do not uniquely identify TCL-derived waters, but they do provide a useful indicator for shallow compared to deep ground-water flow paths and general rock/water interaction times. Information in this report provides a comparison between the geochemistry within the TCL and in the region as a whole.

Introduction

Near Tuba City, Arizona (fig. 1), the Tuba City Landfill (TCL, fig. 2) operated as an unregulated and unsupervised waste-disposal site starting in the 1940s. After the initiation of environmental monitoring by the Bureau of Indian Affairs (BIA) in 1995, radionuclides were discovered in the shallow TCL ground water at levels exceeding the U.S. Environmental Protection Agency (USEPA) maximum contaminant levels. The TCL was administratively closed in 1997. A detailed discussion of the TCL history and local hydrogeology can be found in Morgan (2002).

Purpose and Scope

In 2006, the BIA contracted with the U.S. Geological Survey (USGS) to better understand the source of radionuclides in the ground water at the Tuba City Landfill compared to the surrounding region. Uranium deposits are distributed naturally at levels of economic importance in the surrounding area. Ores derived from the Chinle Formation (fig. 2) were processed in the nearby Rare Metals mill

(RMM, fig. 2). It is important to note that no uranium deposits have been reported in the bedrock at the TCL site. In March 2006, geochemical data were collected by the USGS and Walker and Associates, Inc., from five TCL wells and from various regional rocks, sediments, ground water, springs, and surface water (figs. 3–5). This report summarizes those data and presents interpretations that focus on a comparison of the geochemistry in the rocks and water from the Tuba City region. This summary was originally released to the BIA as an administrative report that was not publicly available.

Table 1 summarizes the number of samples collected, the analyses performed, and the laboratories that produced the data. These data include whole rock and sediment samples that were collected and analyzed to determine metal and selected isotopic composition. Rocks and sediments were leached to determine the potential for these solids to release metals and other elements into the ground water. Ground water and surface water were collected from wells, springs, and streams to determine the regional geochemistry and compare it to the geochemistry within the TCL, including isotopic differences. A master database of analyses is provided in Appendix A, and all of the original data provided by the laboratories are in Appendix B. For simplicity, the TCL master database (Appendix A) does not include analyses that were consistently below detection limits, removes analysis overlap (elements determined by multiple methods), organizes samples by geologic units, and orders the analyses for easier comparisons.

Regional Hydrogeologic Setting

This section is provided as background information on the hydrogeology of the region based on previous studies. Additional hydrogeologic details on a smaller scale, along with new information provided by this study, are provided in the "Results and Discussion" section. Both the TCL and the Rare Metals mill (RMM) are located east and, on a regional scale, across-gradient from Tuba City (TCL and RMM, fig. 6). The principal aquifer beneath the TCL and RMM is the weakly cemented and highly permeable Navajo Sandstone, which is often referred to as the N-aquifer. Near Tuba City, springs emerge along the contact between the Navajo and underlying Kayenta Formation in Pasture Canyon and Moenkopi Wash. The Navajo Sandstone and Kayenta Formation interfinger near the TCL; thus, the mapped geologic contact between these formations is not considered precise (fig. 2). Near the landfill, the transition zone between the Navajo and Kayenta consists of alternating mudstone, siltstone, and sandstone, which likely has a local influence on shallow ground-water flow paths. Surficial sediment consists of windblown eolian sand and silt, which may be transported long distances. Dune features are common, and surficial eolian deposits are more permeable than the underlying bedrock. Additional details on the hydrogeology of the region are in Morgan (2002) and Cooley and others (1969). Additional hydrologic and geochemical data relating to the hydrogeology of the region are in Lopes and Hoffman (1996) and Truini and Macy (2007).

The direction of ground-water movement near the TCL is west to southwest toward Pasture Canyon (fig. 6), about 1 mile from springs that emerge within the Hopi village of Moencopi. Near RMM, the direction of ground-water movement is southward toward Moenkopi Wash (fig. 6). The regional water table in figure 6 is based on spring elevations provided in USGS topographic maps of the area, which were digitized on top of the regional geology from Cooley and others (1969). Detailed ground-water flow gradients on a local scale are beyond the scope of this regional report. Downgradient from the TCL and RMM, springs and streamflow in Pasture Canyon and Moenkopi Wash are used for irrigation, livestock, and domestic drinking water.

Most uranium ore processed at the RMM was derived, not from Navajo Sandstone near Tuba City, but from ores originating west and downgradient from Tuba City. A major source was the Chinle Formation near Cameron, Arizona. Some uranium has been found in the lower Kayenta and also along the lower contact of the Chinle with the underlying Moenkopi Formation near Cameron. Uranium ores also came from the Orphan Lode mine, a breccia-pipe structure in the Redwall Limestone (on the south

rim of the Grand Canyon) and from the Monument Valley area, as well as from other Atomic Energy Commission (AEC) buying stations (*http://www.eia.doe.gov/cneaf/nuclear/page/umtra/tubacity_title1.htm*).

Analytical Methods

The analytical methods used for this study are similar to those used in a study of the upper Verde River headwaters (Wirt and DeWitt, 2004). Details on these methods are found on page E6 of that report, which also provides a background discussion on using isotope characterization and apparent age-determination techniques (pages E6–E8). In brief, samples for water analyses were analyzed using inductively coupled plasma–mass spectrometry (ICP–MS) (Lamothe and others, 2002), inductively coupled plasma–atomic emission spectrometry (ICP–AES) (Briggs, 2002), ion chromatography (IC) (Theodorakos and others, 2002) and were titrated in the laboratory for alkalinity. Water samples for ICP–MS and ICP–AES analyses were filtered in the field using a 0.45-micrometer capsule filter and acidified to a pH < 2 with ultrapure nitric acid. Water samples for IC and alkalinity also were field filtered to 0.45 micrometers and were refrigerated for preservation. Samples analyzed for ^{18}O, ^{2}H, ^{3}H, ^{13}C, ^{14}C, ^{34}S, ^{87}Sr, and $^{234}U/^{238}U$ ratios were not filtered unless suspended sediment was apparent, and these samples were not acidified. Table 1 summarizes the laboratories that completed the isotopic analyses. Uranium concentrations also were measured at Northern Arizona University (NAU), and these independent uranium concentration values are considered more accurate than the ICP–MS analyses completed at the USGS (P.J. Lamothe, U.S. Geological Survey, oral commun., 2006). All plots of uranium concentrations in water use the NAU values. Whole rock and sediment samples were crushed and then digested with acid for elemental analyses using ICP–MS. Rock and sediment leachates were obtained by crushing the sample to pass a < 100 mesh (0.149 mm) screen, and 50 g of material was placed in a 1-L bottle with deionized (DI) water (20 to 1 leaching ratio). The solid sample was in contact with the leachate and shaken horizontally for 18 hours, after which water samples were extracted with a syringe and filtered to 0.45 micrometers. The leachates were preserved using the same procedures previously discussed for ICP–MS and ICP–AES analyses.

Results and Discussion

Geology and Radioactivity

The data for this study must be considered in a three-dimensional setting. As such, the regional geochemistry data are presented in a graphical information system (GIS) along with cross sections and graphs. The regional geology of the area is provided in figure 2. The term "regional" is used in the context of the area shown in figure 2, where the TCL and the RMM locations are labeled. In the vicinity of the TCL, the geologic formations with increasing depth are the Navajo Sandstone, Kayenta, Moenave, and Chinle Formations with starting depths of approximately 0, 300, 800, and 1,100 feet, respectively. Details on the geology of these formations are discussed in detail by Morgan (2002) and Cooley and others (1969). For this report, the important geologic features (1) are generally flat-lying formations, and (2) have a general sequence of grain size becoming finer with depth, where ground-water flow in the Chinle Formation is likely very limited near Tuba City. In the Navajo, Kayenta, and Moenave, finer layers exist and occur in greater abundance with depth, which may locally act as confining units for ground-water flow. As a result, most of the ground-water flow is in the Navajo Sandstone and Kayenta Formation, and the ground-water flow direction generally follows topography. Much of the uranium-bearing ore for the nearby RMM was from the Chinle Formation (fig. 2). In general, original exploration data done in 1954 by the USGS (under contract to the Atomic Energy Commission) did show radiation above the average background in areas within the Chinle Formation

and radiation below background in areas within the Moenave, and Kayenta Formations, and Navajo Sandstone (Morgan, 2002). However, the data from the 1954 study did not cover the Tuba City region (airborne radiometrics and soil data, R. Kucks and P. Hill, U.S. Geological Survey, oral commun., 2007).

Radiometric coverage of the whole United States for the NURE project was conducted from 1974 through 1983 with various flight-line intervals. In the Tuba City area (part of the Marble Canyon quadrangle) the flight lines had 3-mile spacings in the east-west direction with north-south tie lines at various spacings (fig. 7). The airborne radiometric NURE data from this time period has been recompiled and is available in Duval (1999) and the original data from Marble Canyon is in LK&B Resources, Inc. (1980). A second phase of NURE work in the area was completed in 1979 and 1980 (Field and Blauvelt, 1982), which included geologic field work and soil data collection (no data collection in the Tuba City area, R. Kucks, U.S. Geological Survey, oral commun., 2007). The report by Field and Blauvelt (1982) summarizes the additional geologic data for the NURE project and specifically states that favorable environments for uranium deposits exist in the Chinle Formation, whereas the Navajo Sandstone, Kayenta, and Moenave Formations are unfavorable environments for uranium deposits.

The airborne radiometric data for Marble Canyon are provided in figure 8 as a contoured image of the original data using the east-west flight lines. The radiometric data are converted from measured radioactivity to a uranium concentration of surficial materials. For reference, the average crustal abundance for uranium is 2.3 ppm (Fortescue, 1992). In figure 8, the outline of the Chinle Formation shows higher uranium concentrations (5 ppm) compared to the area north of Tuba City with uranium concentrations (< 1.6 ppm) below the average crustal abundance. A closer image of the Tuba City area compared with the geology (fig. 9) shows the decrease in uranium concentrations throughout most of the Navajo Sandstone compared to the much higher uranium concentrations in the Chinle Formation. An anomalous area of high uranium concentrations (> 5.8 ppm) is northeast of the RMM tailings site and is centered on and just south of Greasewood Lake (figs. 10 and 11). The source of this anomalously high uranium concentration at the surface is unknown but may be related to the evaporation of surface water in Greasewood Lake. While figures 8–10 provide general trends in uranium concentrations, the area actually being measured by the airborne instrument is limited to approximately 100 to 150 meters on either side of the flight line (A. McCafferty, U.S. Geological Survey, oral commun., 2007). The width of the flight lines in figure 10 represents the actual area of ground measurement, which shows the TCL and the RMM tailings ponds were not directly measured by the NURE radiometrics. As a result, the NURE data are useful for regional trends in uranium concentrations but cannot be used for very local, site-scale interpretations.

Additional radiometric data from the USEPA Navajo Abandoned Uranium Mines Project (completed in the Tuba City area in 1999) show little radiation above background levels (pink color indicates detections above average background) in the areas outside of the Chinle Formation, except for an area to the east of the tailings ponds near the RMM site and south of Greasewood Lake (figs. 11 and 12). This area is probably related to the area of anomalously high uranium seen in the NURE data (fig. 10). As seen in figures 11 and 12, the flight areas for the USEPA project only covered limited portions of the Tuba City area and are not nearly as extensive as the flight areas in the previously discussed NURE data.

Ground-Water Flow and General Geochemistry

The regional water table generally follows the topography, with flow toward Moenkopi Wash and locally near the TCL site toward Pasture Canyon (fig. 6). Cross section A–B in figure 6 locates the conceptual cross section shown in figure 13, where the light blue shading indicates the saturated zone, and the top of the water table generally follows the topography. Conceptual flow lines show how ground

water follows shallow or deep flow paths depending on the topography. Shallow flow lines have shorter travel paths and less rock/water interaction. Deeper flow lines indicate ground water that has probably recharged outside of the defined "regional" study area with very long travel paths and significant rock/water interaction times. In addition, ground-water flow in the Moenave Formation probably has a smaller flow rate and may only discharge to surface water in the most western portions of the region due to its greater depth below Tuba City. Ground-water flow in the Chinle Formation below the TCL is probably insignificant due to its low permeability. Sampling of these different flow paths with depth can be achieved using multiple-level ground-water sampling techniques. While some wells in the TCL have been installed more deeply (approximately 100 to 200 ft), these depths are still only within the shallow portion of the Kayenta Formation. As a substitute for multilevel well data, flow paths are identified from geochemical and age-determination techniques in spring water. Unfortunately, spring samples near Moenkopi Wash and Pasture Canyon may be a mix of shallow and deeper ground waters, as these are areas where flow lines converge (fig. 13).

All of the 25 water samples were modeled in PHREEQC (Parkhurst and Appelo, 1999) using the MINTEQ database (Allison and others, 1990) in order to include arsenic and uranium. The PHREEQC input and output files are included in Appendix C. This modeling is a good check on the analytical chemistry to double check charge balances (all waters should be neutrally charged) and to compare the water chemistry with the interpreted geology. Of the 25 water samples, all except three samples were within a 5-percent charge balance error. The samples outside of this range were 06TCSP109, 06TCSP113, and 06TCSP116 with charge balance errors of 7.6, –9.2 and –21.0 percent, respectively. Saturation indices of the water samples are consistent with water derived from carbonate-cemented sandstones. Most major minerals are undersaturated (not likely to precipitate) with the exception of calcite, aragonite, barite, and chalcedony with median saturation indices of 0.05, –0.09, 0.41, and 0.05, respectively (all near equilibrium values). These minerals are commonly found in carbonate-cemented sandstones and tend to control the concentrations of calcium, carbonate, barium, and silica. The monitoring wells within the TCL have an overall geochemistry that is different from the regional ground waters (that is, greater calcium, strontium, chloride, nitrate, and alkalinity concentrations), which is probably derived from the landfill waste and processes related to the decay of organic carbon (which increases carbon dioxide, lowers the pH with carbonic acid, and increases carbonate solubility). However, the focus of this report is the variation of uranium and other trace-element concentrations.

Metal and Uranium Concentrations in Rock, Leachate, and Water Samples

Sites for regional rock and sediment samples with the appropriate sample identifications (ID) that match Appendixes A and B are in figure 4. Rock and sediment samples from the Chinle Formation are considered to be outside of the Tuba City region, and the site where they were collected are shown in figure 5. Sites for all of the regional water samples are shown in figure 3. Sample IDs of SW indicate surface water, GW and MW indicate ground water, and SP indicates spring water. Whole rock element concentrations (fig. 14) compare the median values (along with the 5th and 95th percentiles) for elements in the Chinle Formation compared to the regional rock samples in the Navajo Sandstone and Kayenta Formation. In order to keep the figure legible, the 5th and 95th percentiles for the Navajo/Kayenta were not included. Only elements that were greater than 100 ppm in the Chinle Formation are plotted. This comparison plot shows the two data sets have the same general trend, and the elements in greatest concentration are Al, Ca, Fe, K, Mg, and Na. The greatest differences in concentration can be seen as the difference in median values or the gaps between sample plotting in figure 14. The greatest differences, in order, are U, As, Co, Pb, and Zn.

A similar element comparison is done for the four sampled TCL wells compared to the 21 regional water samples (fig. 15). Landfill well MW–13D is included in the regional samples because it is a deep well that is not affected by ground water from the TCL. Again, the 5th and 95th percentiles for

the Navajo/Kayenta samples are not shown to maintain legibility and only elements that were greater than 100 ppm in the Chinle Formation rock samples (plus sulfate) are plotted. Figure 15 shows a similar trend in elemental concentrations in water (compared to the element concentrations in the solid phase, fig. 14) with the highest concentrations being Ca, K, Mg, Na, Sr, and SO_4. However, the water concentrations reflect element mobility, such as the low solubility of aluminum and iron in oxidized, high-pH ground waters. The greatest differences in the median concentrations, in order, are Co, Pb, Zn, As, U, and SO_4.

A convenient way to compare elemental concentrations in the Chinle rocks to the Navajo/Kayenta rocks and the TCL ground water to regional waters is to use the ratio of the median concentrations (that is, median cobalt value in the Chinle rocks divided by the median cobalt value in the Navajo/Kayenta rocks). The constituents with the greatest ratios are As, Co, Pb, U, and Zn (fig. 16). The elements with the top five ratios in median values are the same when comparing the rock samples (Chinle and Navajo/Kayenta) and the water samples (TCL and regional), but in a different order of decreasing ratio values (fig. 16). The differences in order for these ratios are probably due to variations in mobility along with the possibility of additional sources in the TCL from the municipal solid waste. Sulfate was included in figure 16 because sulfuric acid was used in the RMM to leach the uranium ore. However, sulfate can also be derived from gypsum and (or) pyrite oxidation in natural rock formations. These data suggest that the source of some metals and sulfate in the TCL ground water may be related to nearby mining and milling operations if the original source material was the Chinle Formation. Only arsenic and uranium are above primary drinking-water standards (USEPA Maximum Contaminant Level for As is 10 ppb and for U is 30 ppb; *http://www.epa.gov/safewater/contaminants/index.html*) in the median value for TCL wells. Cobalt occurs only in concentrations less than 5 ppb, and lead and sulfate can come from a variety of sources (lead may be found in municipal solid waste, for example, car batteries). Given other element mobility and source uncertainties, uranium is used as a "focus" element that is unique to geology and is mobile in shallow, oxygenated ground water.

Uranium concentrations in the solid phase of Chinle rocks, Chinle sediments, regional rocks, and regional sediments are shown in figure 17. Chinle uranium-ore rocks show a clear elevation of uranium concentrations (fig. 17). The lower concentration of uranium in the Chinle sediments reflects the mobility of uranium in oxidizing environments and indicates much of the uranium has been leached away. The regional rocks and sediments have uranium concentrations similar to or slightly less than the average crustal abundance of uranium (2.3 ppm), which is consistent with the NURE radiometric data. Figure 18 shows the areal distribution of uranium concentrations in the rocks and sediments. For regional samples, the highest solid-phase uranium concentrations occur in an evaporite deposit (06TCEV115C, U=2.06 ppm) where evaporation probably concentrated uranium, and in a rock sample that is lower down in the stratigraphic section in the Kayenta Formation (06TCRK115B, U=1.87 ppm). Associated sediments in the area have similar uranium concentrations (06TCSS115A, U=1.17 ppm and 06TCSS102A, U=2.06 ppm). These samples are much lower in uranium concentration than the median uranium concentration in the Chinle rock samples (291 ppm).

A summary of the uranium concentrations in the solid-phase leachates compared to the TCL ground water and regional water shows a distinct similarity to uranium concentration for the Chinle rock leachates and the TCL ground water (fig. 19). Uranium concentrations in leachates from the regional rocks and regional sediments are variable, yet very low in uranium, with no concentrations greater than 2.7 ppb. The Chinle sediment leachates are similar in uranium concentrations (median U=4.3 ppb) to the regional water samples (median U=5.2 ppb), whereas the regional rock and sediment leachates (median U=0.10 ppb and U=0.94 ppb, respectively) have lower uranium concentrations than the regional water samples. The Chinle Formation is located to the west of the TCL, and the prevailing winds in the area are from the west-southwest. Windblown sediment from the Chinle Formation could provide a source of uranium in the regional waters. Other possible explanations of why the regional waters are slightly

higher in uranium concentration than the regional rock and sediment leachates include different real-world weathering scenarios contrasted with laboratory leaching conditions and (or) locally elevated uranium concentrations in the regional rocks that were not sampled. A map of the uranium concentrations in leachates is shown in figure 20. Overall, the data in Appendix A indicate that the highest uranium concentration in the leachates corresponds to rocks or sediments with the highest concentration of uranium.

Details on the uranium concentration in water (regional and TCL) are shown in figures 21 through 23. Figure 21 compares the regional waters with the TCL ground water, where each sampled well concentration is given. This figure shows that most of the TCL ground water has uranium concentrations (median U=35.5 ppb) above the regional concentration (median U=5.2 ppb). The highest uranium concentration is 175 ppb in MW–7. Map views of the regional concentrations are given in figure 22 (wide view) and figure 23 (enlarged view). The nearest upgradient water sample (06TCSP107) has a uranium concentration of 12.5 ppb (figs. 3 and 23), which may be above the regional median concentration due to water evaporation in this hand-dug well where many elements tend to be enriched (the oxygen and deuterium isotopes in this well, as discussed later, also indicate an evaporative influence). The highest regional ground-water uranium concentration occurs in 06TCSP101 (figs. 3 and 22, Goldtooth Spring) with a concentration of 41 ppb. This spring occurs below an area of windblown sand, and the elevated uranium concentration could be from the windblown sand and (or) evaporative effects on the water recharging in this area (this sample did have an anomalously high specific conductance, indicating a high concentration of dissolved solids).

Uranium Isotope Ratios

Uranium isotope ratios ($^{234}U/^{238}U$) were measured with the hope that these ratios could differentiate TCL ground water from regional ground water and possibly assist in determining the uranium source. Uranium isotope ratios in TCL wells are uniquely low compared to the regional samples (figs. 24 through 27). The high median uranium isotope ratio tends to identify the regional waters (fig. 25), but a low uranium isotope ratio cannot be used to uniquely distinguish TCL landfill-derived water from regional water due to the large range in uranium isotope ratios for the regional water (fig. 25).

Uranium isotope ratios reflect rock/water contact time. Short contact times, such as those re-created using the rock and sediment leachates, should produce uranium isotope ratios close to one, presuming the uranium source rock has not been subject to major oxidative leaching within the last million years (Zielinski and others, 1997). This is seen for the Chinle rock and sediment leachates, with slightly higher ratios for the regional rock and sediment leachates (fig. 24). Longer rock/water interaction times, such as seen in the regional ground water, should produce much higher ratios, with typical ground water $^{234}U/^{238}U$ ratios having a range of 1–3 in the Tuba City region (figs. 25 and 26). These higher ratios occur because isotopic fractionation creates excess dissolved ^{234}U during prolonged, mild leaching of uranium by ground water (Zielinski and others, 1997).

The definition of “longer” as opposed to “shorter” contact times is being investigated through the quantitative derivations of chemical weathering and fluid flow rates (Maher and others, 2006). For this study, longer contact times are considered to be thousands of years, and shorter contact times are several hundred years or less. Longer contact time produces ratios around 2 to 3 as seen in the regional waters (figs. 24 and 25). Ground water in the TCL presumably has had shorter rock/water interaction time given the age of the landfill (<60 years) and shows a median ratio of 1.73 (figs. 24 and 25). This ratio most likely represents mixed ground waters as ground water that is less than 60 years old from the landfill (uranium isotope ratio near 1) and incoming regional ground water that is older than 60 years (uranium isotope ratios around 2 to 3). In nearby regional waters on the south side of Moenkopi Wash (figs. 3 and 26), the uranium isotope ratios for 06TCSP101 and 06TCSP102 are 1.69 and 1.84,

respectively. These and other regional springwater samples with shorter rock/water interaction times may contain a "younger" component of ground water and show uranium isotope ratios less than 2 (figs. 26 and 27). This makes the use of uranium isotope ratios most applicable to identifying rock/water interaction time and not a direct identifier of ground water from the TCL.

Oxygen and Deuterium Isotopes

The use of ^{18}O and ^{2}H (deuterium) are helpful in determining ground-water recharge conditions. In general, water recharged under cooler climatic conditions is more depleted in ^{18}O and ^{2}H, which has been observed in the nearby Black Mesa area (Truini and Longsworth, 2003). Isotope fractionation also occurs during evaporation, which produces an enrichment of ^{18}O and ^{2}H (Coplen, 1993). Due to the location of Arizona within the continent, ground-water samples appear to fall upon a local water line (LWL) that is approximately parallel to the global meteoric water line (fig. 28). In addition, evaporation during recharge and (or) evaporation from the top of the water table can produce a water line with a lower slope (Coplen, 1993). This may be the case for the shallow TCL wells and for 06TCSP107 (fig. 28), which is a shallow, hand-dug well that could have experienced evaporation due to the open nature of the well and (or) through shallow water table evaporation. Much of the other regional waters appear to have experienced little to no evaporation and fall along the local water line (fig. 28). The separation of ^{18}O and ^{2}H concentrations along the LWL does not follow any consistent trends to differentiate waters that recharged under unique climatic conditions, except for the TCL wells and 06TCST107, which appear to have experienced some evaporation (green line in figure 28).

Tritium and ^{14}C Isotopes

Sampling of tritium (^{3}H) and ^{14}C was completed for water age determination. Tritium was released into the Earth's atmosphere in large quantities during nuclear bomb testing in the 1950s. The current level of tritium in the atmosphere in the southwestern United States is approximately 5 tritium units (TU) (A. Manning, U.S. Geological Survey, oral commun., 2007). A surface-water sample in Pasture Canyon (06TCSW118) had a tritium concentration of 4.9 TU, which probably represents current exchange with the atmosphere. Water samples with values of tritium below detection limit and (or) < 1 TU are mainly pre-bomb water (pre-1952). Samples with 1 to 2 TU may have a significant portion of pre-bomb water but also may have some post-bomb water due to mixing. Except for the Pasture Canyon sample, the highest tritium concentration in the regional waters is 2.3 TU. This indicates that some ground water in the region is less than 60 years old, but a significant portion of the ground water is older than 60 years. The tritium concentration in MW–6 is 445 TU and 16.3 TU in MW–7. MW–6 is located within the TCL and MW–7 is just downgradient. Both of these tritium values are greater than any regional waters and appear to indicate the presence of tritiated water (less than 60 years old) within and just downgradient from the TCL.

Analyses of ^{14}C are often used to date waters in the 500–50,000-year range. As meteoric water is recharged to the ground water, carbon is incorporated as it travels through the soil and generally has a value of post-modern carbon (pMC) on the order of 85 to 95 percent, but a value of pMC greater than 80 percent can still have an age less than 1,000 years (A. Manning, U.S. Geological Survey, oral commun., 2007). While exact age determination using ^{14}C requires the understanding of all carbon sources from the soil, carbonate rocks, and anthropogenic sources, general values without detailed modeling are useful for approximate ages. Only four ^{14}C samples were analyzed for this study (figs. 29 and 30). The values of pMC greater than 80 percent are likely less than 1,000 years old, and the large pMC concentration at MW–7 (108.5) may be due to carbon sources within the landfill and (or) to post-bomb pulse water (which generally has ^{14}C values greater than 100 pMC). The pMC for ^{14}C of 60.7 percent in 06TCSP103, Susungva Spring (figs. 3, 29, and 30), indicates the water source for this spring is likely several thousands of years old.

^{13}C, ^{87}Sr, and ^{34}S Isotopes

Data were also collected for ^{13}C (water and leachates), ^{87}Sr (rocks only), and ^{34}S isotopes (selected water and leachates) and are provided in Appendixes A and B. These isotopes were collected to test whether or not they have the potential to assist in identifying ground-water flow paths. Analyses of ^{13}C isotopes in ground water appear to be consistent with recharge through a soil zone resulting in some carbonate dissolution. ^{87}Sr isotopes show only a small variation in rock samples, and water samples were not analyzed for ^{87}Sr. ^{34}S isotopes are quite variable in rock samples and have the potential to identify sulfur sources from gypsum compared to pyrite. The ^{34}S variation in water shows some potential for identifying unique ground-water flow paths, but no consistent trends were identified. Because the ground-water flow paths contact multiple sulfur sources such as gypsum, pyrite, and possibly sulfuric acid from mill processes, additional research would be required to more fully identify the rock/water interactions. Additional interpretations of ^{13}C, ^{87}Sr, and ^{34}S isotopes are beyond the scope of this report.

Summary and Conclusions

This report summarizes data collected for the comparison of geochemistry in the rocks and water from the Tuba City region by the USGS in 2006. Of concern at the TCL is the source of uranium and other elements in the ground water, given the fact that uranium ore was mined from the Chinle Formation (mining area located just west of the TCL, fig. 2) and that uranium ore was processed in the RMM (located just east of the TCL, fig. 2).

A conceptual cross section along line A–B in figure 30 shows locations where analyses of ^{18}O and ^{2}H indicate evaporation, tritium indicates relative ground-water ages, uranium concentrations identify regional and TCL sources, and the uranium isotope ratios indicate rock/water contact time (fig. 31). All of these data are complementary, as water samples in the Tuba City region identify shallow and deep ground-water flow paths as indicated (flow lines in figure 31), with Susungva Spring being the deepest and longest flow path (no evaporation, very low tritium, low uranium concentration, and high uranium isotope ratio). Uranium and tritium concentrations are potential identifiers of ground water flowing through the TCL.

Data from this report indicate that ^{18}O and ^{2}H, tritium, and ^{14}C are good indicators of ground-water flow paths. The hydrogeology of the Tuba City area is relatively simple, with ground-water flow directions following the regional topography. Deeper ground water in the area has longer flow paths and older ground water ages, as this water was recharged many miles upgradient in the higher elevation areas to the northwest. Ground water flowing through the TCL is very shallow, and the landfill waste is barely into the water table. The resulting uranium isotope ratios in ground water at the TCL are likely due to mixing of recent recharge through the TCL (short rock/water contact times of 60 years or less) and upgradient regional ground-water flow (longer rock/water contact times). Similar uranium isotope ratios are seen in the shallow regional ground water, making uranium isotope ratios another indicator of ground-water flow paths, but not a unique tracer for uranium source. Major-element data from rock samples in the Chinle Formation compared to regional rocks show much higher concentrations of U, As, Co, Pb, and Zn (in order of greatest ratios of median values). Likewise, data from the TCL wells and the regional waters show the same set of elevated elements with a different order of greatest median value ratios of Co, Pb, Zn, As, and U. However, the geochemical controls on these elements can be very complex, and the possibility exists that landfill sources other than mill/mine-related material could produce these elevated metal concentrations (such as lead from car batteries). Because of the complexity of looking at various other constituents, uranium is an important element to understand as a geochemical indicator.

Based on the facts that uranium does not occur in significant amounts in the Navajo Sandstone and Kayenta Formation, and that uranium was mined in the Chinle Formation, the uranium detected in

the TCL is potentially mining related. The most compelling evidence that uranium in the ground water in and around the TCL is related to mill/mine wastes from Chinle Formation mining is based on the following observations: (1) uranium in regional rock samples is very low (median, 0.5 ppm) compared to rock samples from the Chinle Formation (median, 291 ppm), (2) uranium in regional waters is very low (median, 5.2 ppb) compared to ground water in the TCL (median, 35.5 ppb), (3) uranium and other metal concentrations in leachates derived from the Chinle Formation are very similar to the concentrations in the TCL ground water, and (4) airborne radiometric surveys close to the Tuba City Landfill do not indicate any radiation values in the immediate Tuba City area above background (high uranium concentrations in the near-surface soils around Greasewood Lake are the exception).

While this report includes only a limited number of TCL wells, the data indicate uranium concentrations in the landfill ground water are above regional concentrations (that is, ground water in the Navajo Sandstone and Kayenta Formation). In addition, the source appears to be closely related to material that was derived from the Chinle Formation, which is known to be a source of ores processed at the nearby RMM. The TCL is a unique geochemical environment where potentially uranium-bearing waste and ground water with elevated uranium concentrations appear to be in direct contact with municipal solid waste. The geochemical controls on uranium mobility in this environment must be understood in order to approach remedial efforts most effectively. Remedial efforts may include containment and (or) remediation of the ground water. Understanding the controls on uranium and other metal mobilities may provide more effective and cost-efficient solutions.

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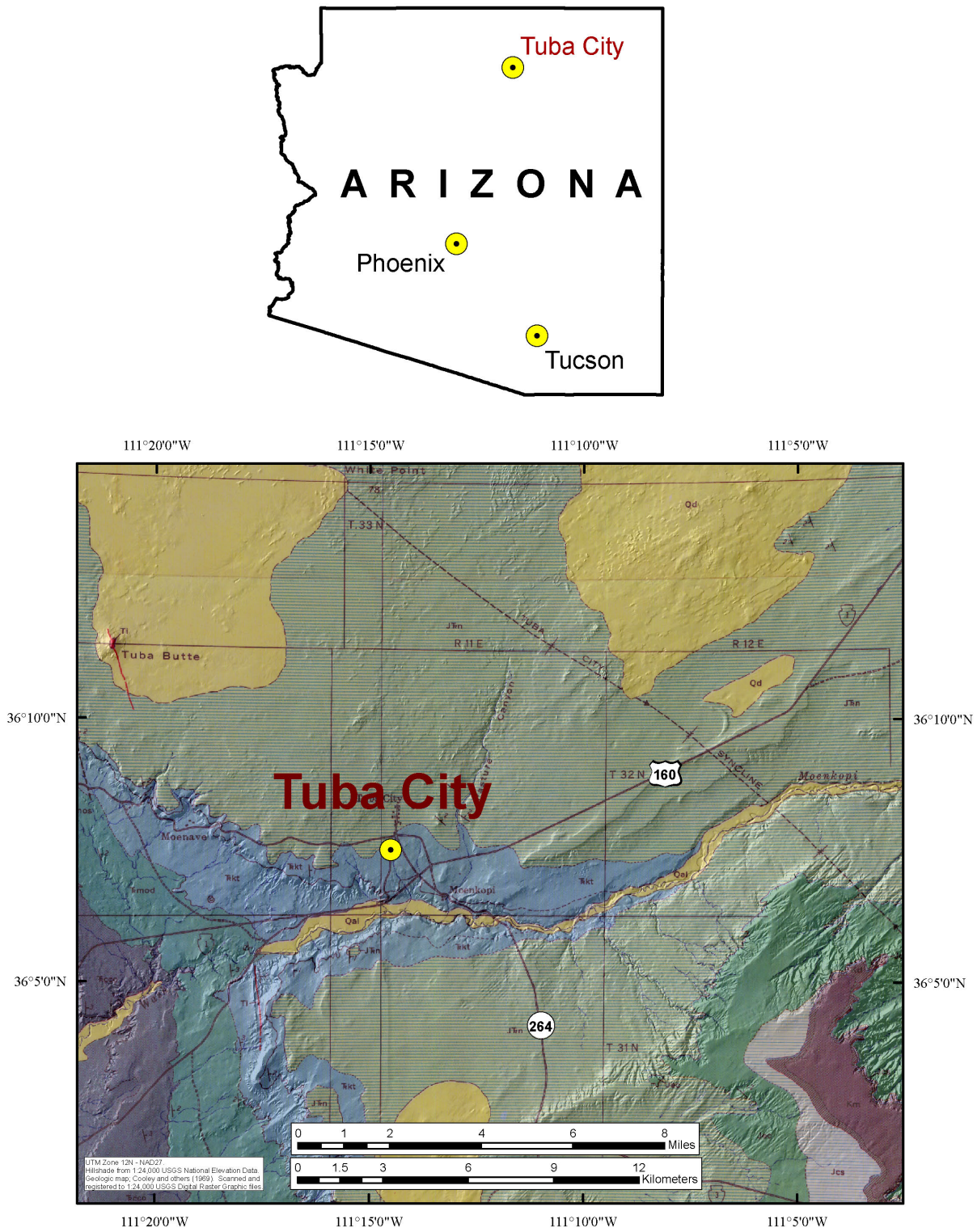

Figure 1. Location map for Tuba City, Arizona.

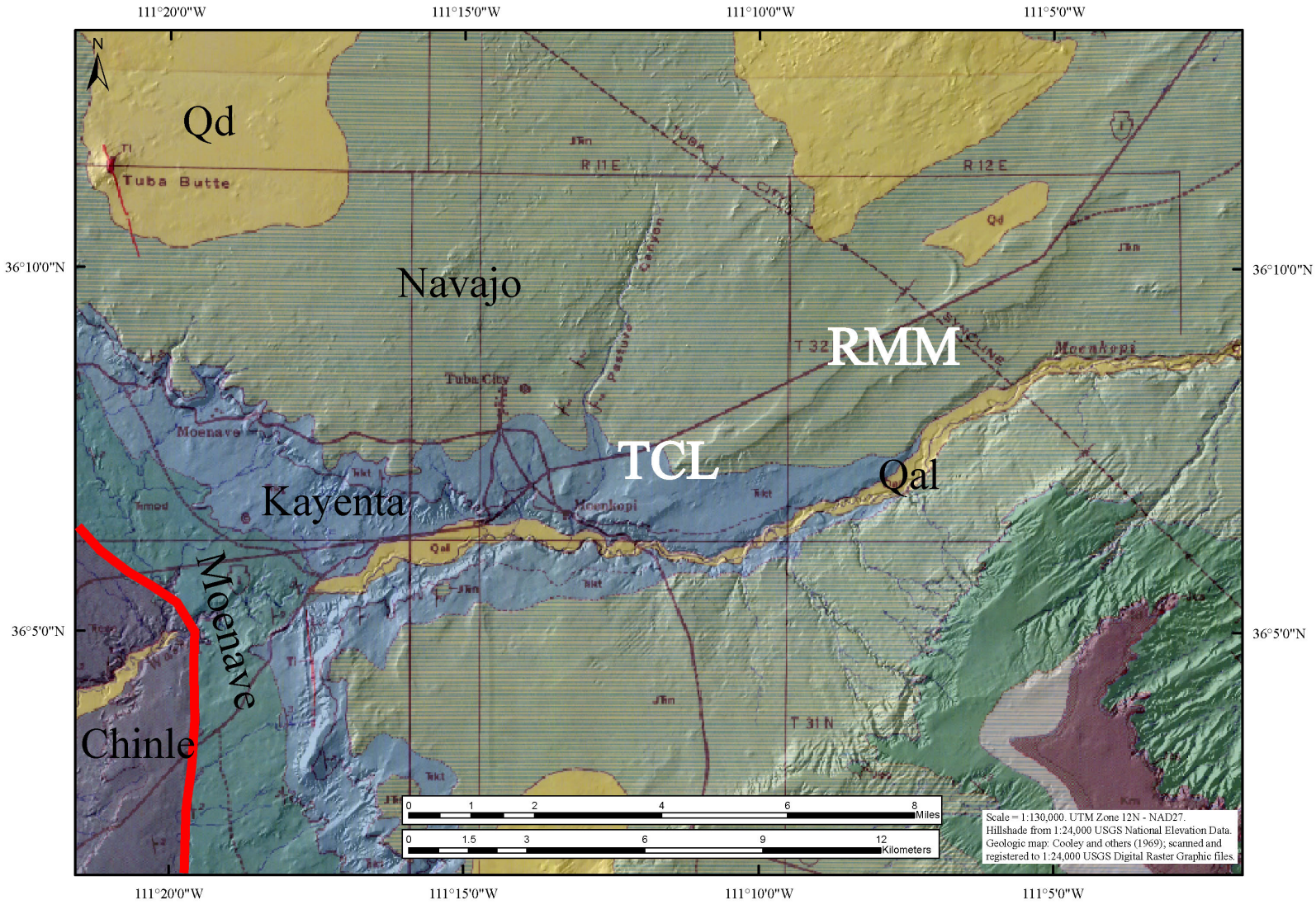

Figure 2. Regional geology around the Tuba City Landfill (TCL). Rare Metals mill site is labeled as RMM. Red line highlights the edge of the Chinle Formation. Qd = dune sand, Qal = alluvium. Geology is from Cooley and others (1969).

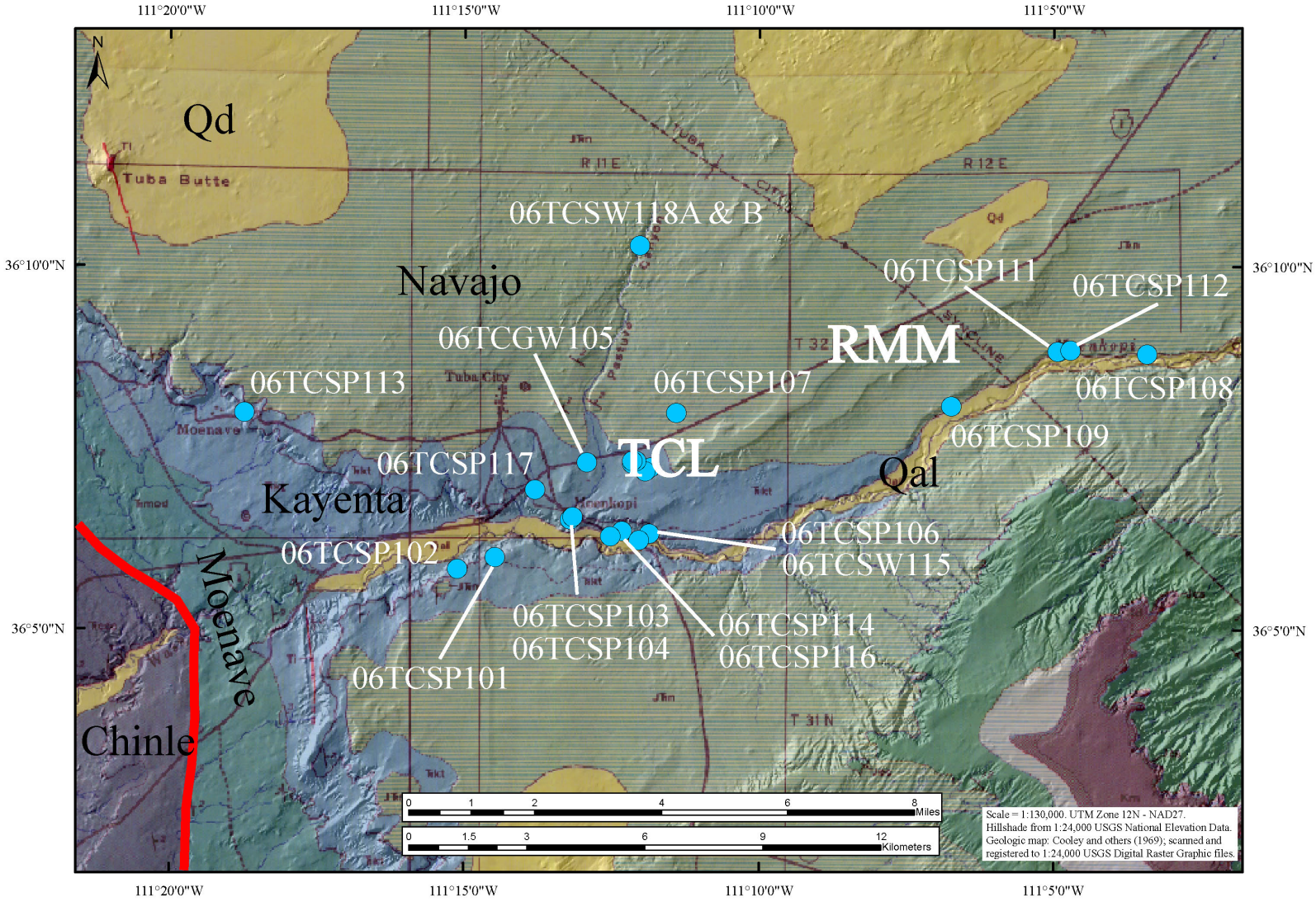

Figure 3. Water-sampling sites. Water samples include ground water, springs, and surface water. SP in sample identification is a spring, GW in sample identification is ground water, and SW in sample identification is surface water. All of these samples are considered "regional." Red line highlights the edge of the Chinle Formation. The TCL wells are all ground water and are labeled in subsequent figures.

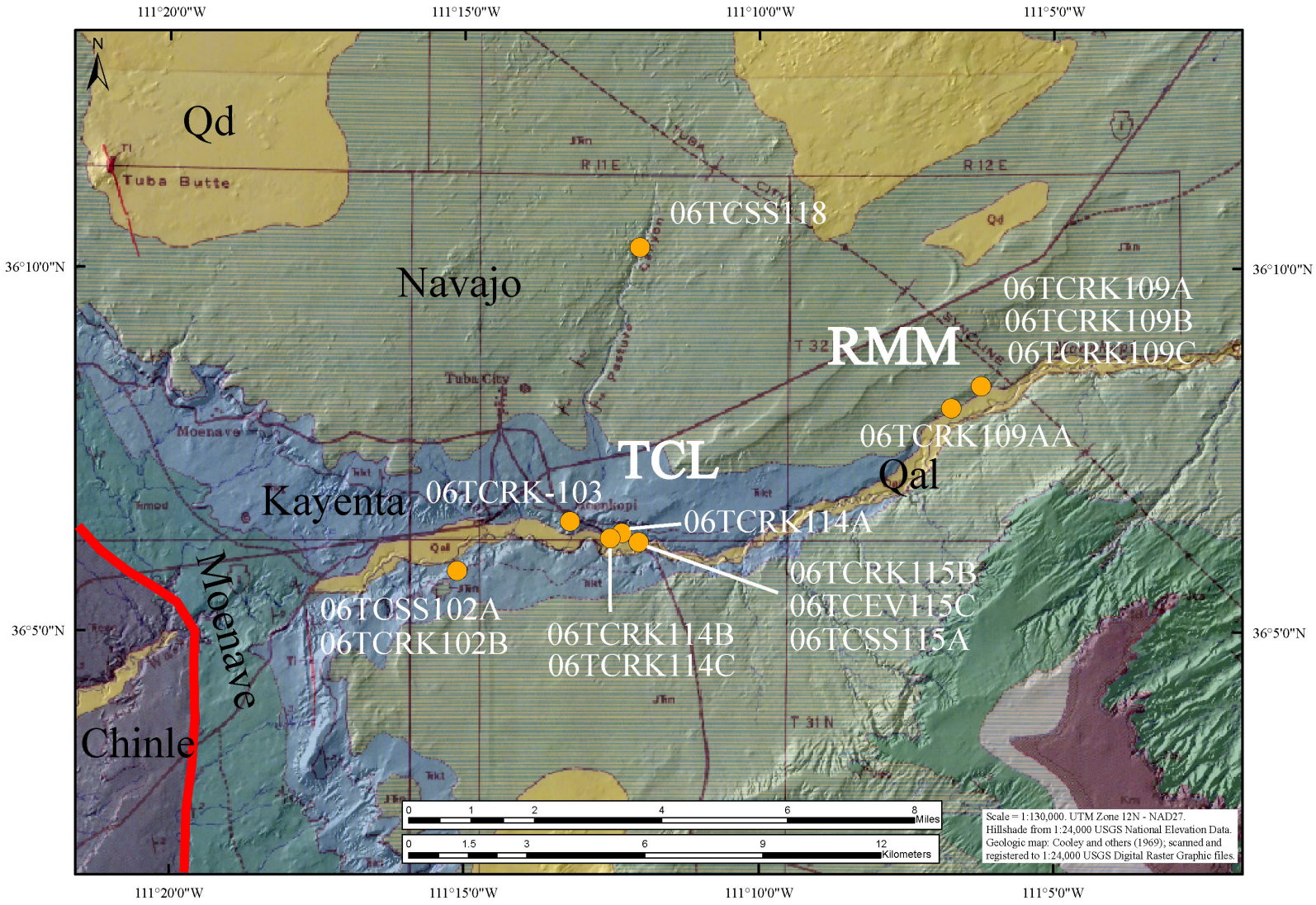

Figure 4. Solid-phase sampling sites. RK in sample identification is a rock sample. SS in sample identification is a sediment sample. All of these samples are considered "regional." The edge of the Chinle Formation is highlighted in red.

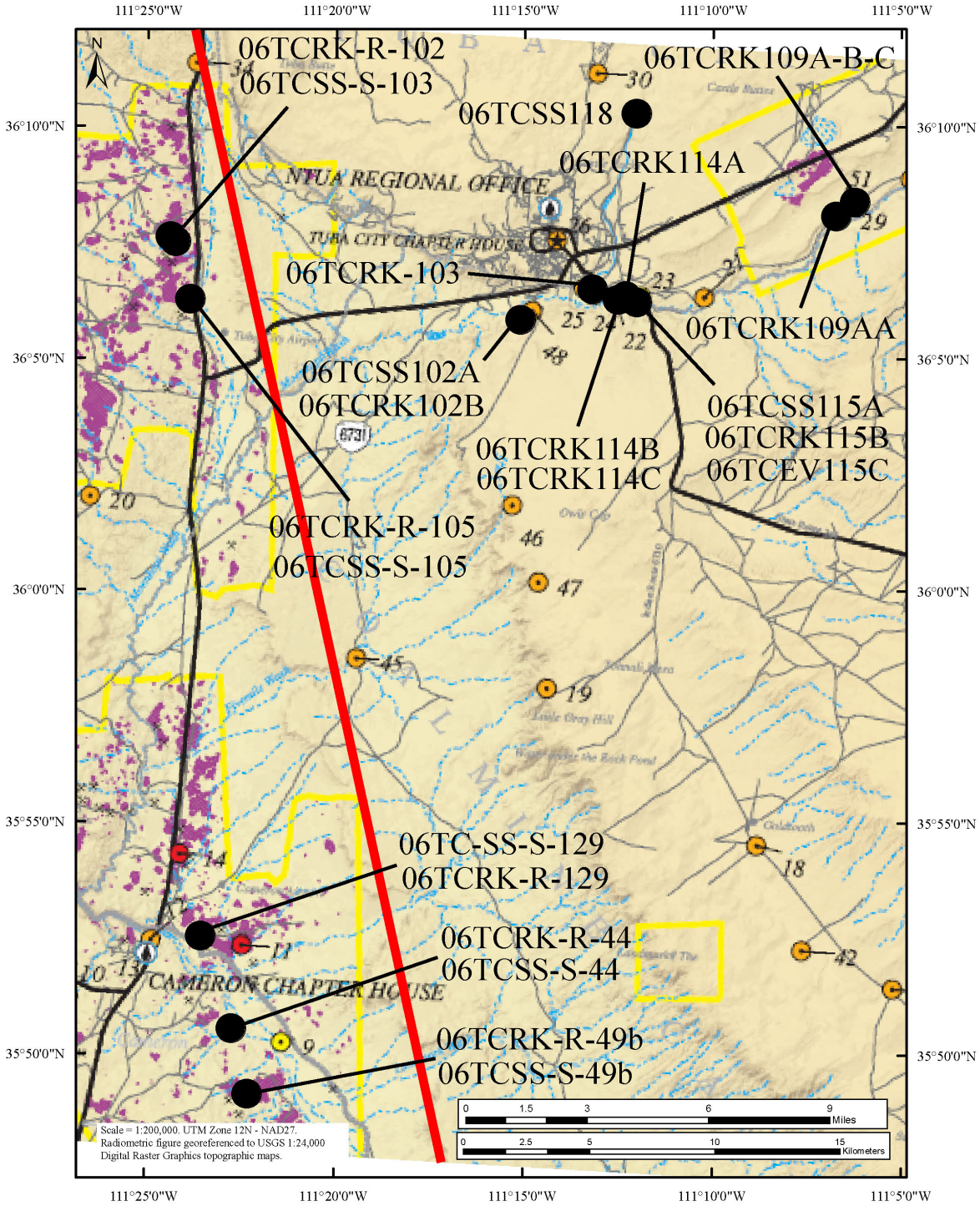

Figure 5. Location of Cameron mine area solid-phase sampling sites in addition to the Tuba City regional solid-phase sampling sites. RK in sample identification is a rock sample. SS in sample identification is a sediment sample. The addition of an R in the rock samples and the addition of an S in the sediment samples identify solid-phase samples taken from mines in the Chinle Formation. The red line highlights the edge of the Chinle Formation. Base map shows radiometric data from the U.S. Environmental Protection Agency Navajo Abandoned Uranium Mines Project. Purple color indicates radiation above background levels, and yellow outline indicates measured areas.

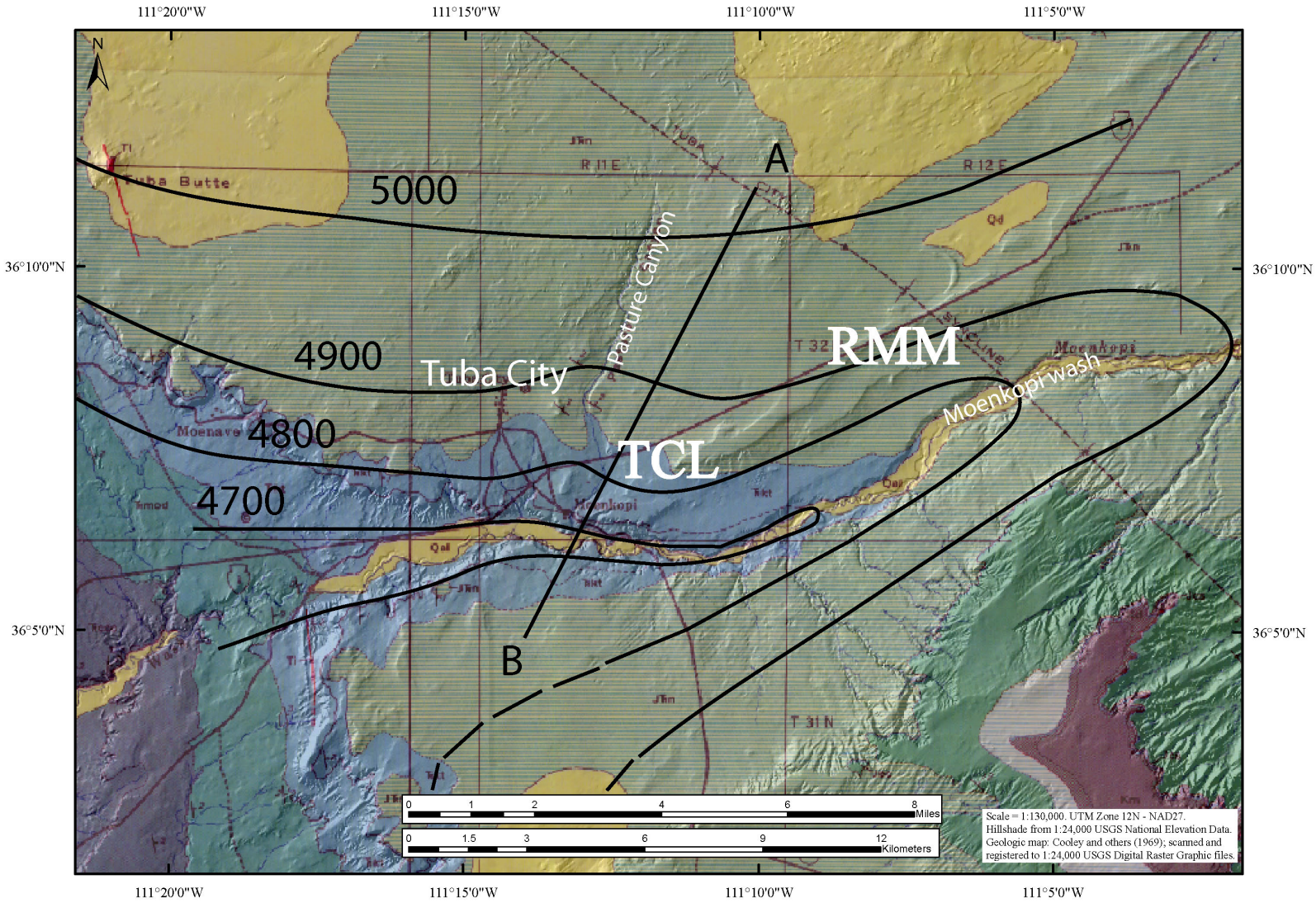

Figure 6. Regional water table overlain on regional geology. Water table is based on spring elevations, which are assumed to measure the top of the water table. Line A–B indicates the location of a conceptual cross section shown in figure 13. Contour interval is 100 ft.

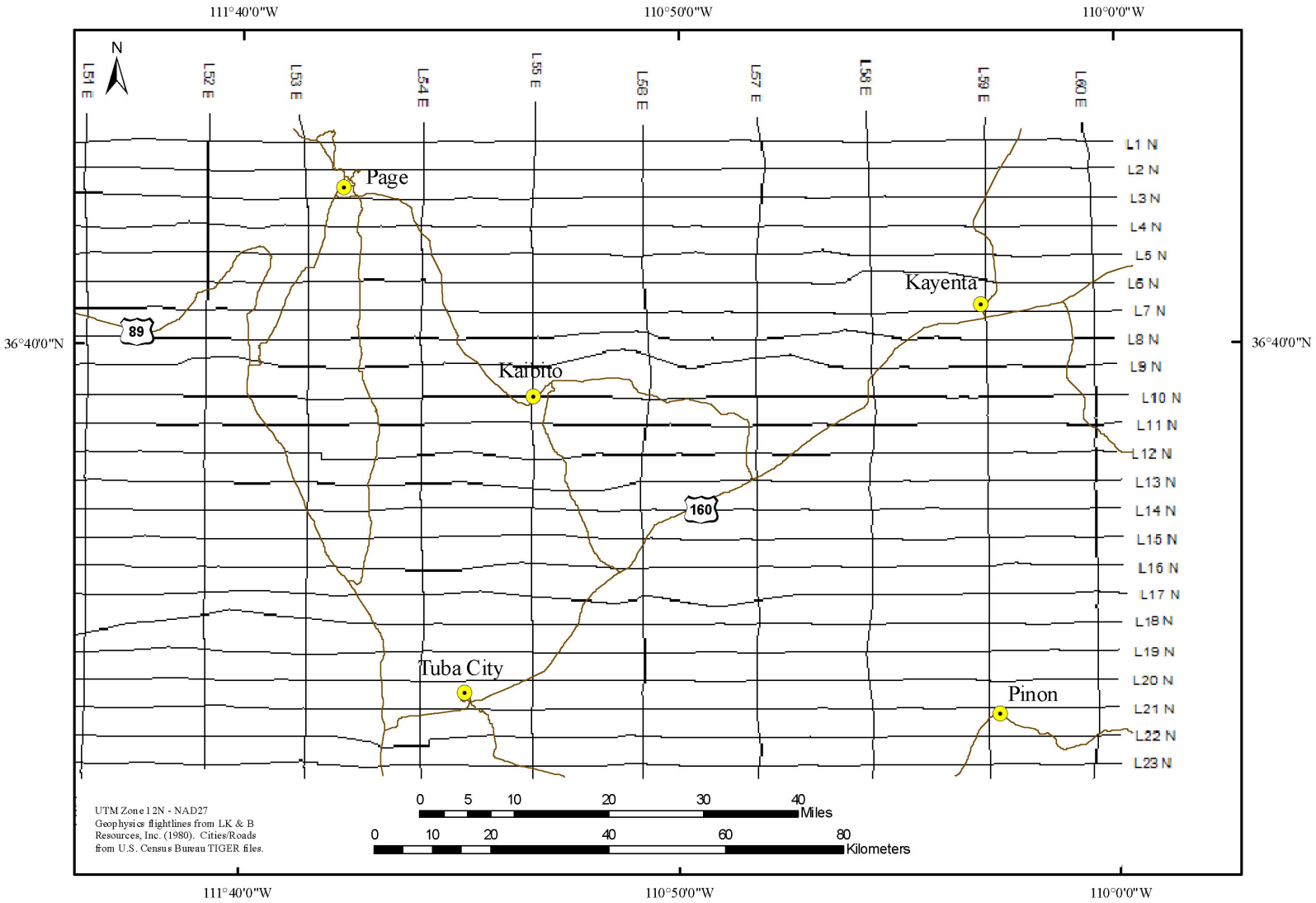

Figure 7. Flight lines for National Uranium Resource Evaluation (NURE) radiometric data in the Marble Canyon quadrangle with 3-mile spacing in the east-west direction.

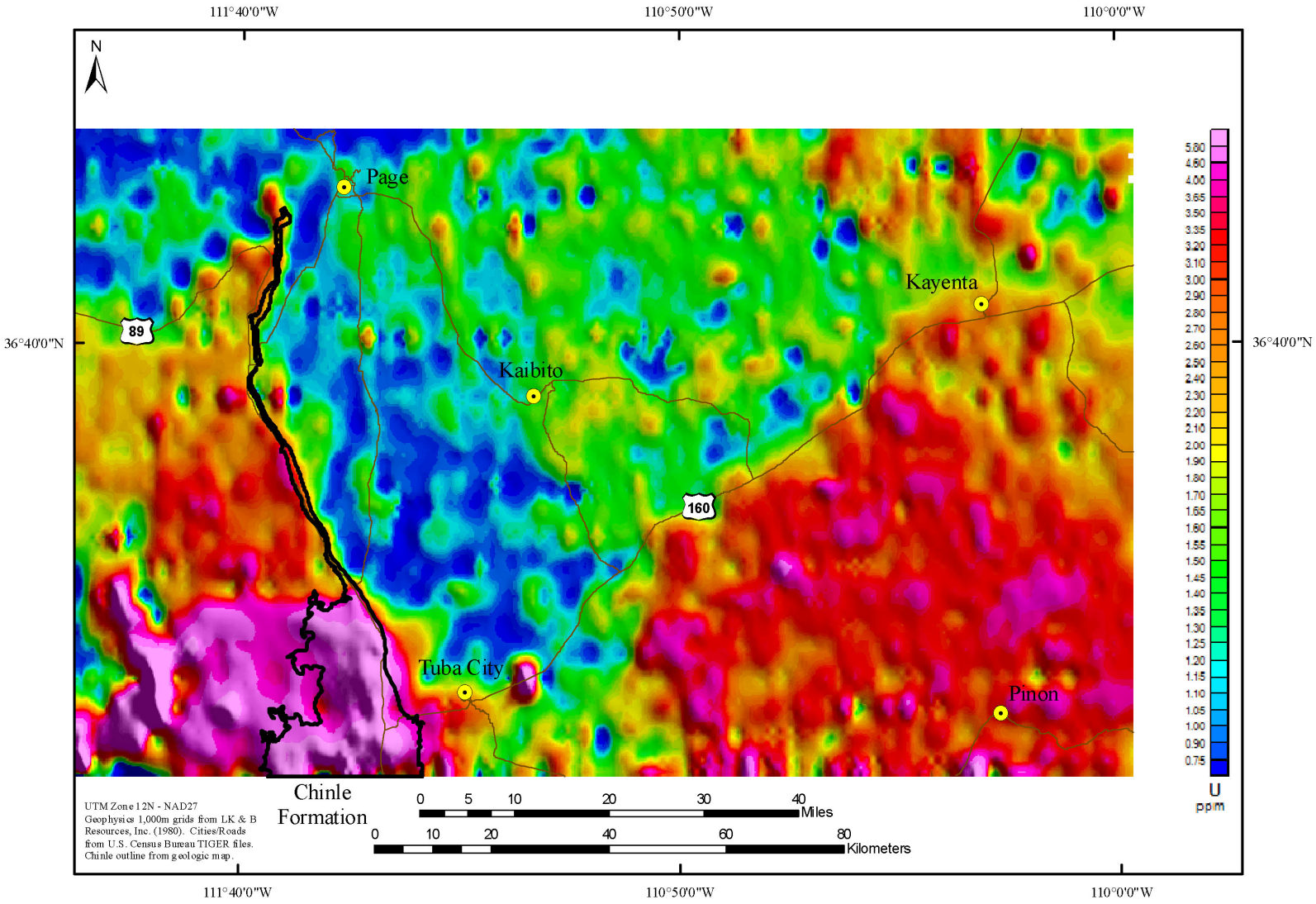

Figure 8. Contoured National Uranium Resource Evaluation (NURE) uranium data for the Marble Canyon quadrangle. Chinle Formation is outlined in black.

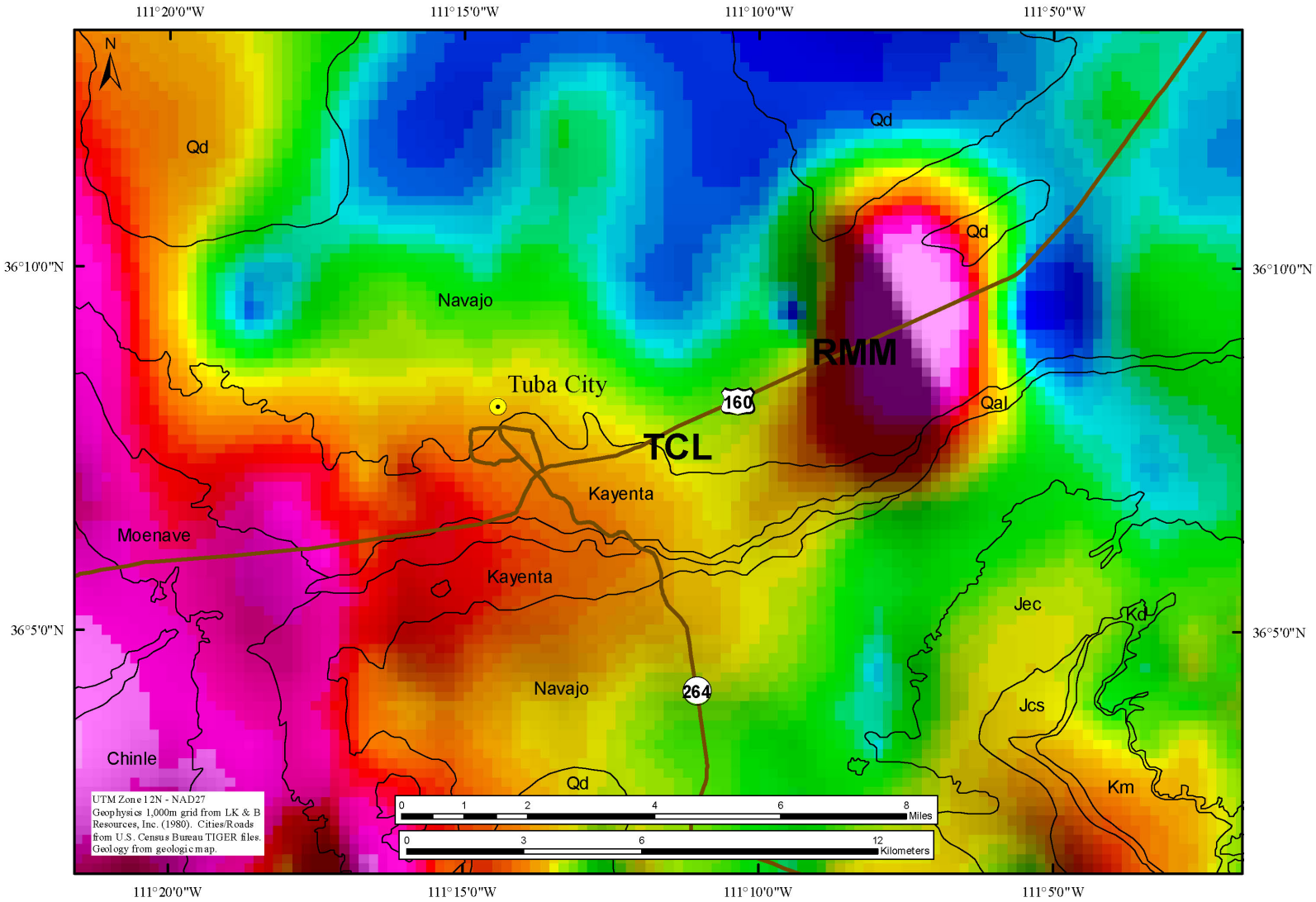

Figure 9. Contoured National Uranium Resource Evaluation (NURE) uranium data for the Tuba City region. Color concentration scale is the same as shown in figure 8. Geologic formations are outlined in black.

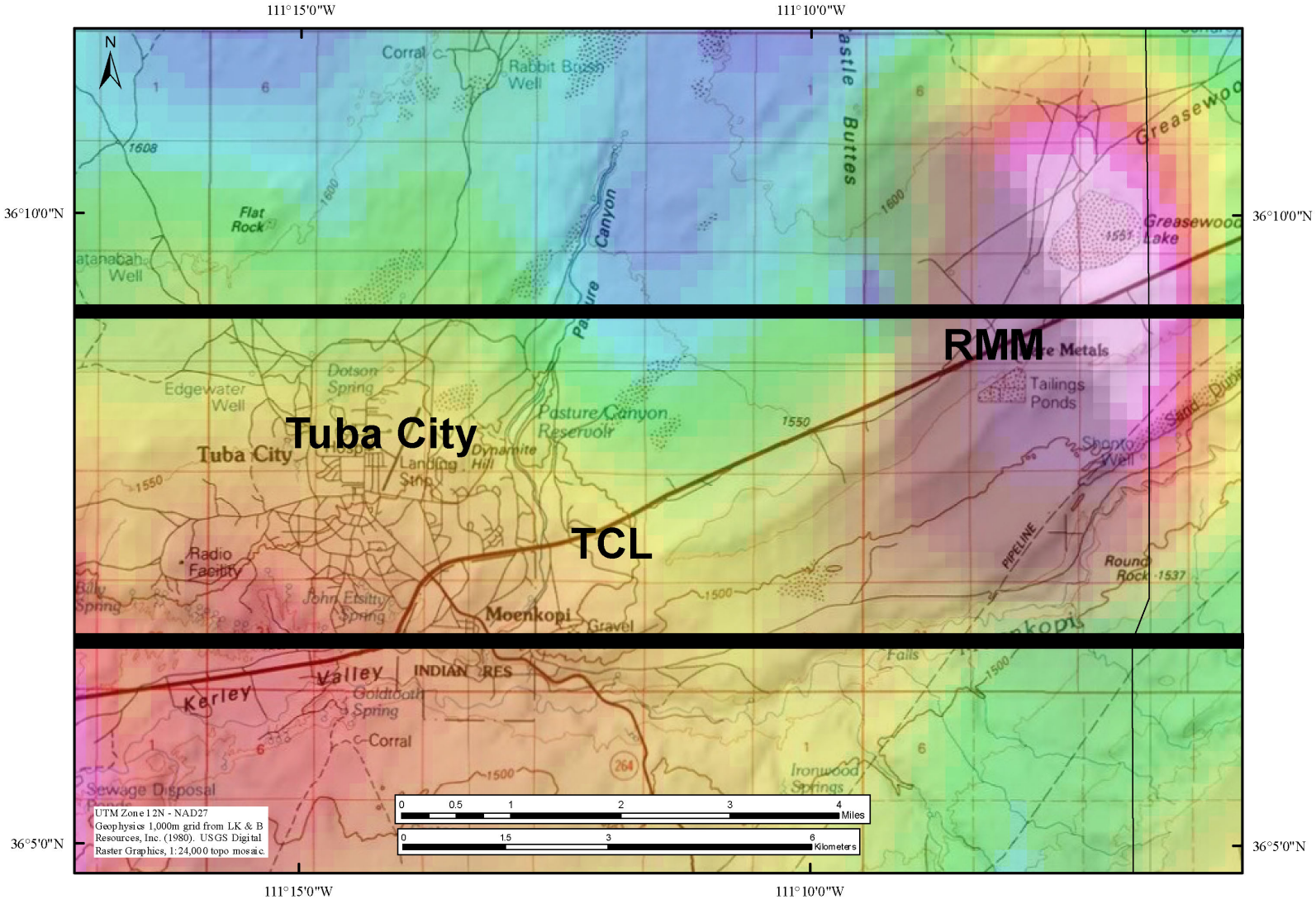

Figure 10. Contoured National Uranium Resource Evaluation (NURE) uranium data with the Tuba City Landfill and the RMM located. Color concentration scale is the same as shown in figure 8. Actual aerial detection areas are given to scale as the thickness of the east-west flight lines.

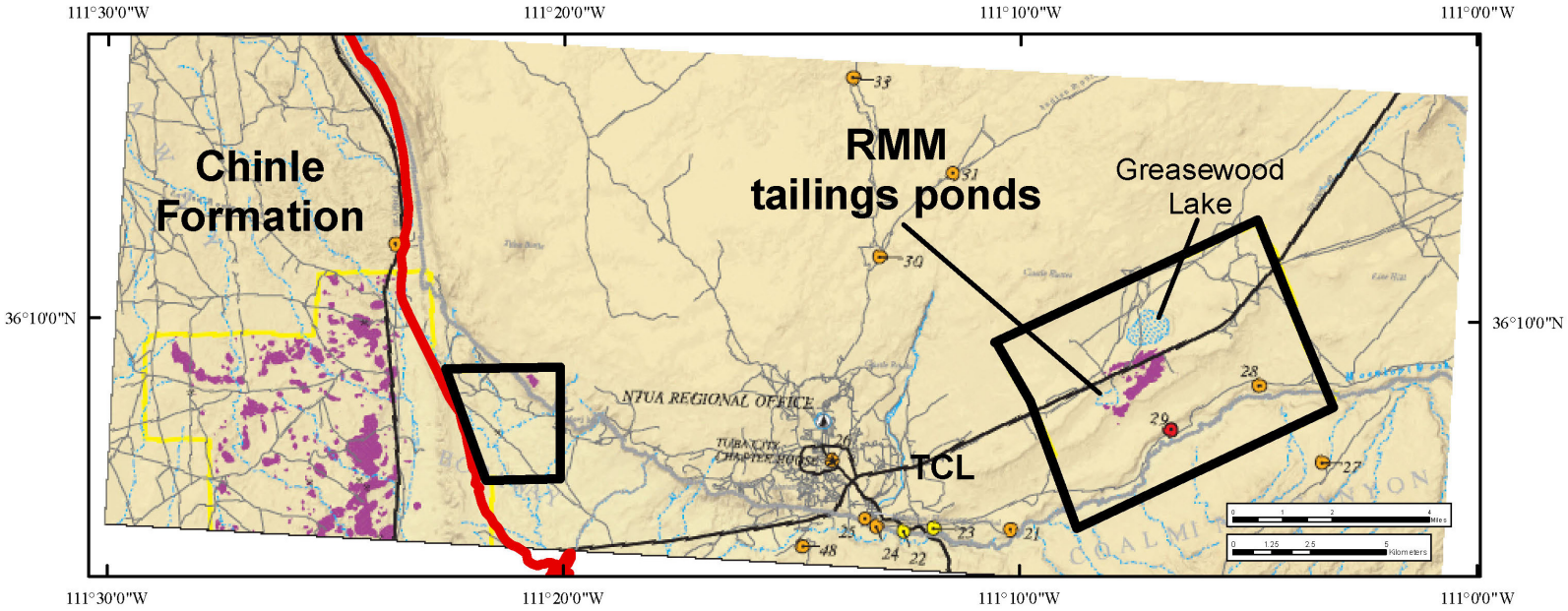

Figure 11. Radiometric data from the U.S. Environmental Protection Agency Navajo Abandoned Uranium Mines Project. Purple color indicates areas with radiation higher than background. The edge of the Chinle Formation is highlighted in red. Areas of radiometric data are outlined in yellow within the Chinle Formation, and areas outside of the Chinle Formation are outlined in black.

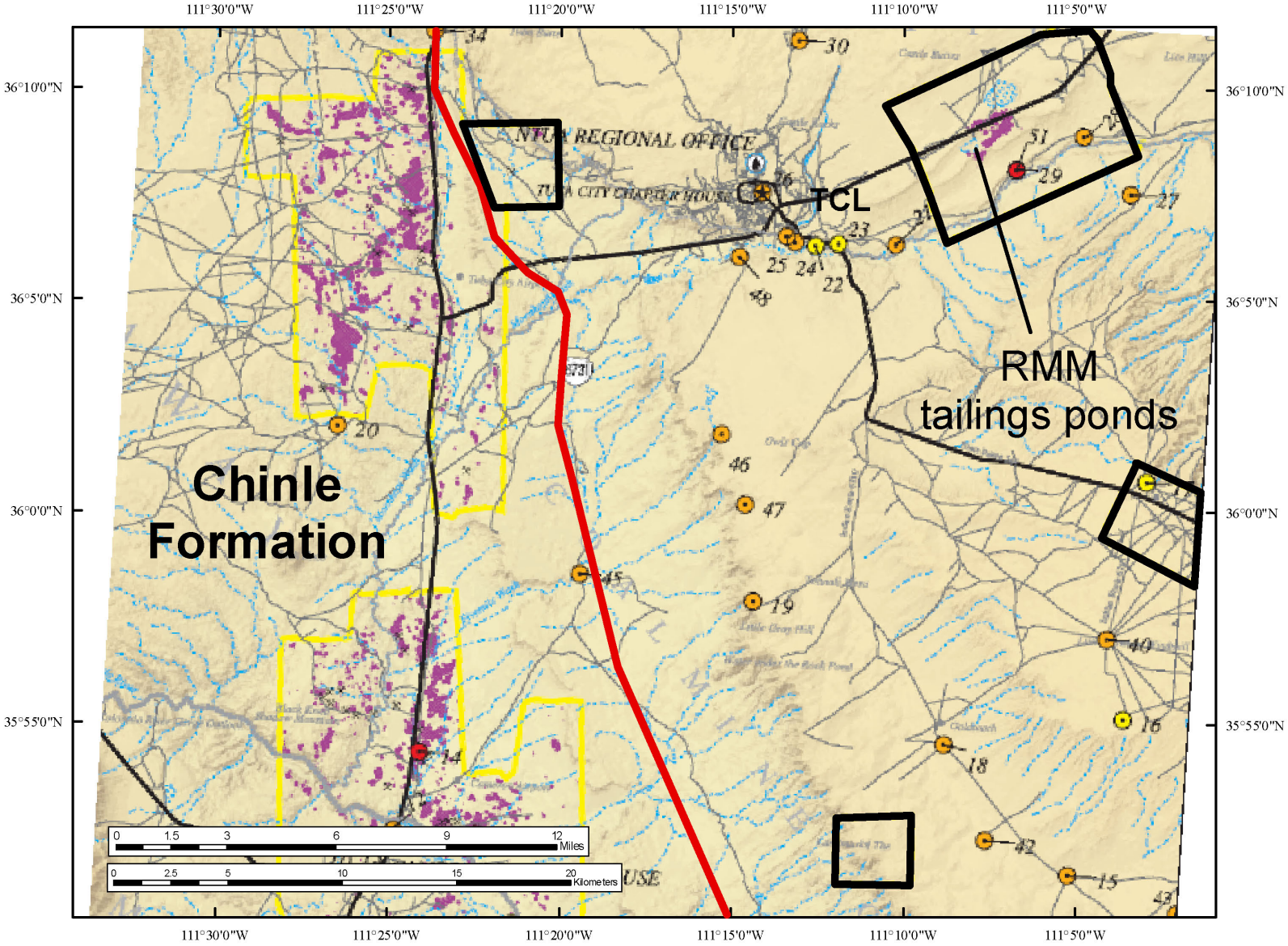

Figure 12. Larger view of the radiometric data from the U.S. Environmental Protection Agency Navajo Abandoned Uranium Mines Project. Purple color indicates areas with radiation that is higher than background. The edge of the Chinle Formation is highlighted in red. Areas of radiometric data are outlined in yellow within the Chinle Formation and areas outside of the Chinle Formation are outlined in black.

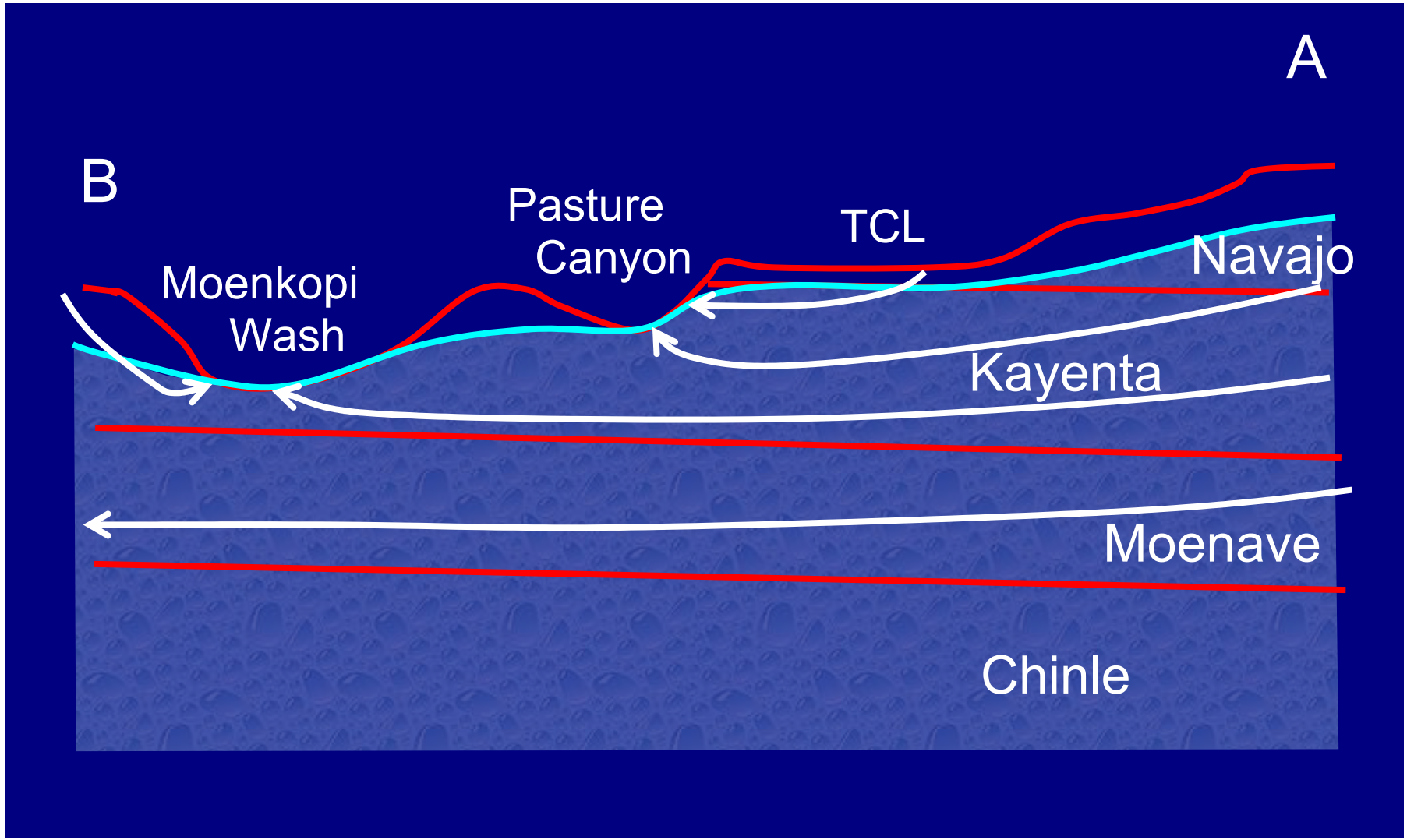

Figure 13. Conceptual cross section along line A–B located in figure 6. Light blue color indicates saturated zone. White lines indicate possible ground-water flow lines.

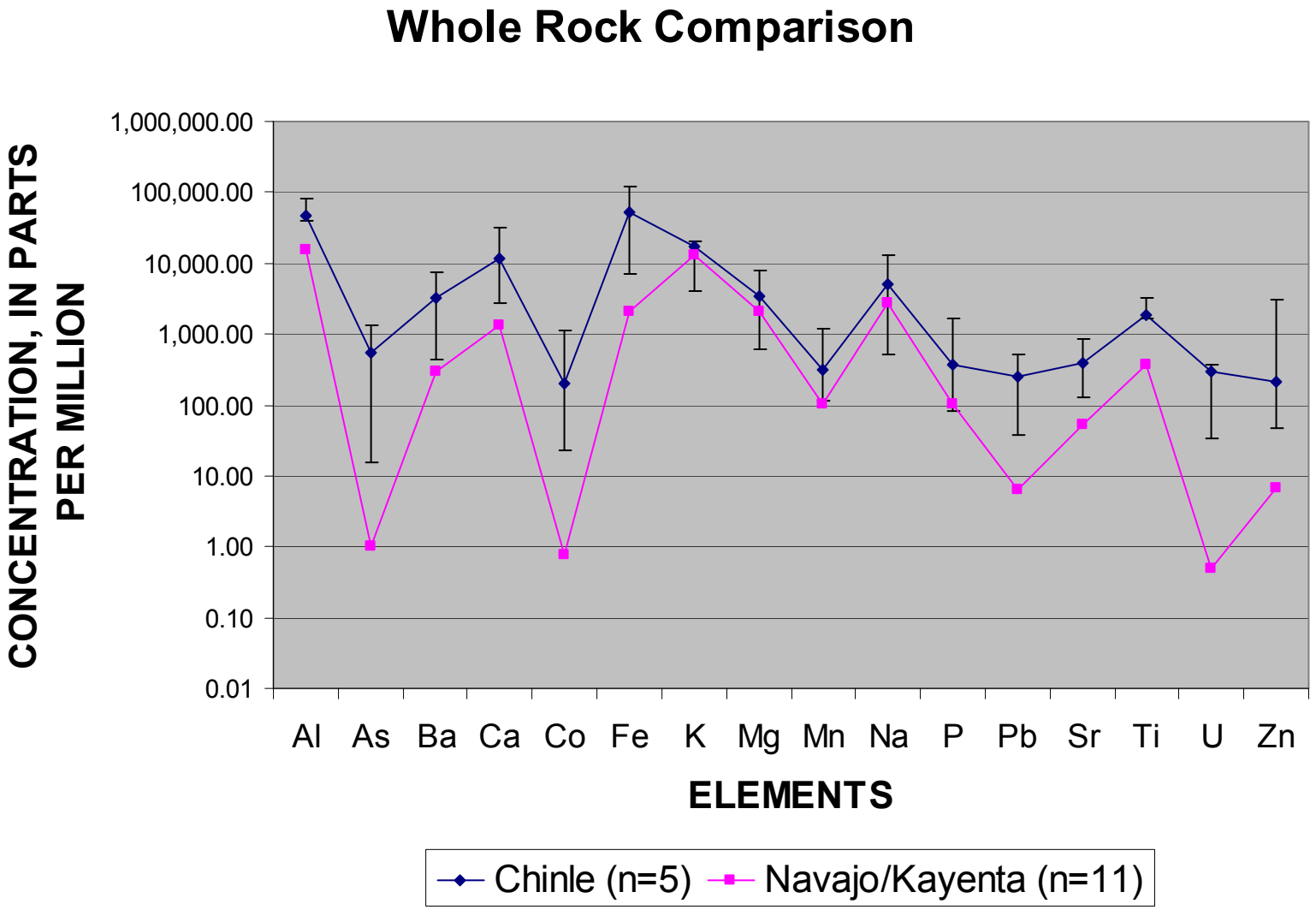

Figure 14. Comparison of element concentrations from whole rock samples in the Chinle Formation compared to the Navajo/Kayenta regional whole rock samples. Only elements that were greater than 100 parts per million in the Chinle Formation are plotted. Plotted points are median values, and "error bars" indicate the 5th and 95th percentiles. The number of samples is indicated (n). Note the log scale for concentrations.

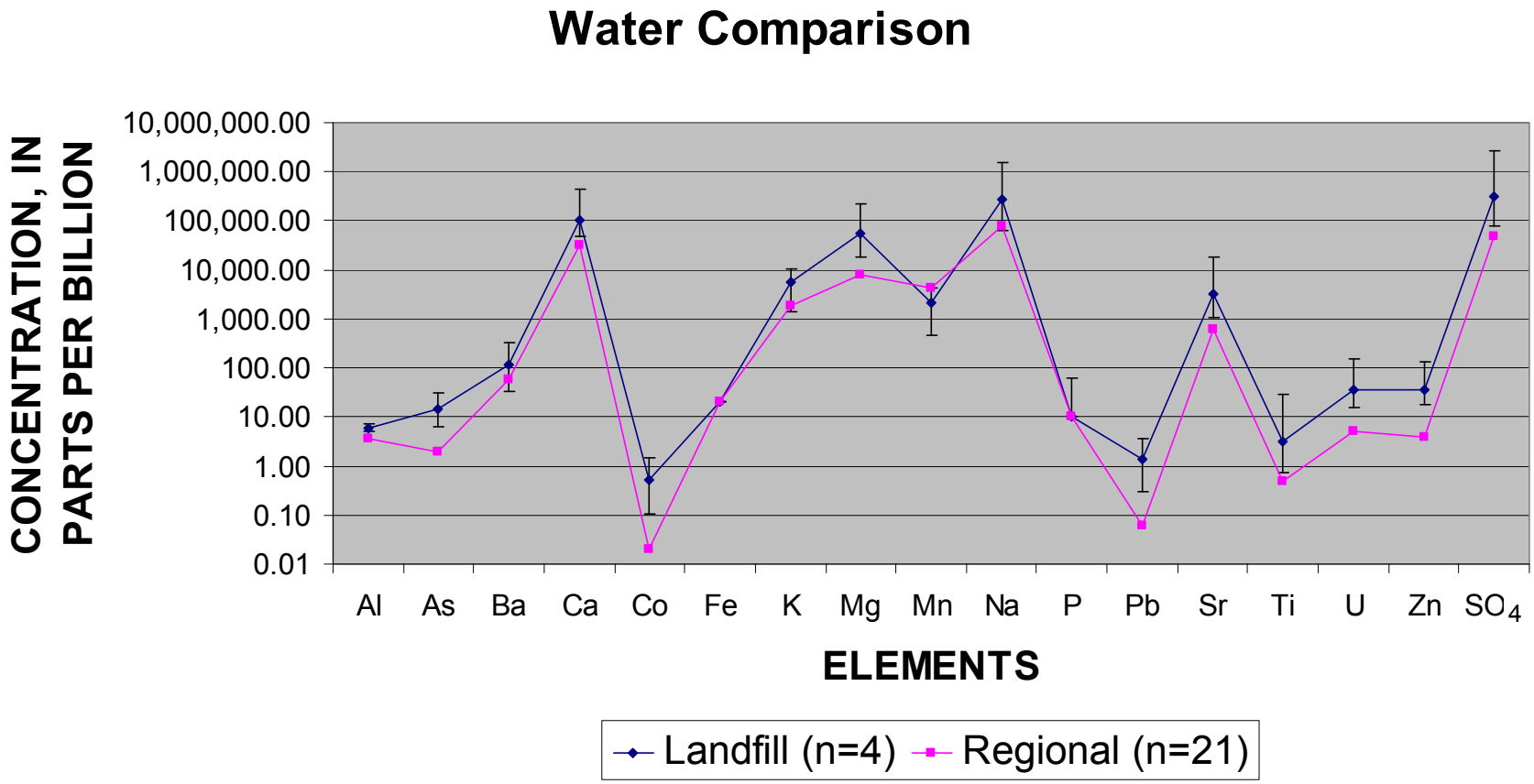

Figure 15. Comparison of water concentrations from the Tuba City Landfill and regional water samples. Only elements that were greater than 100 parts per million in the Chinle Formation (with the addition of sulfate) are plotted. Plotted points are median values and "error bars" indicate the 5th and 95th percentiles. The number of samples is indicated (n). Note the log scale for concentrations.

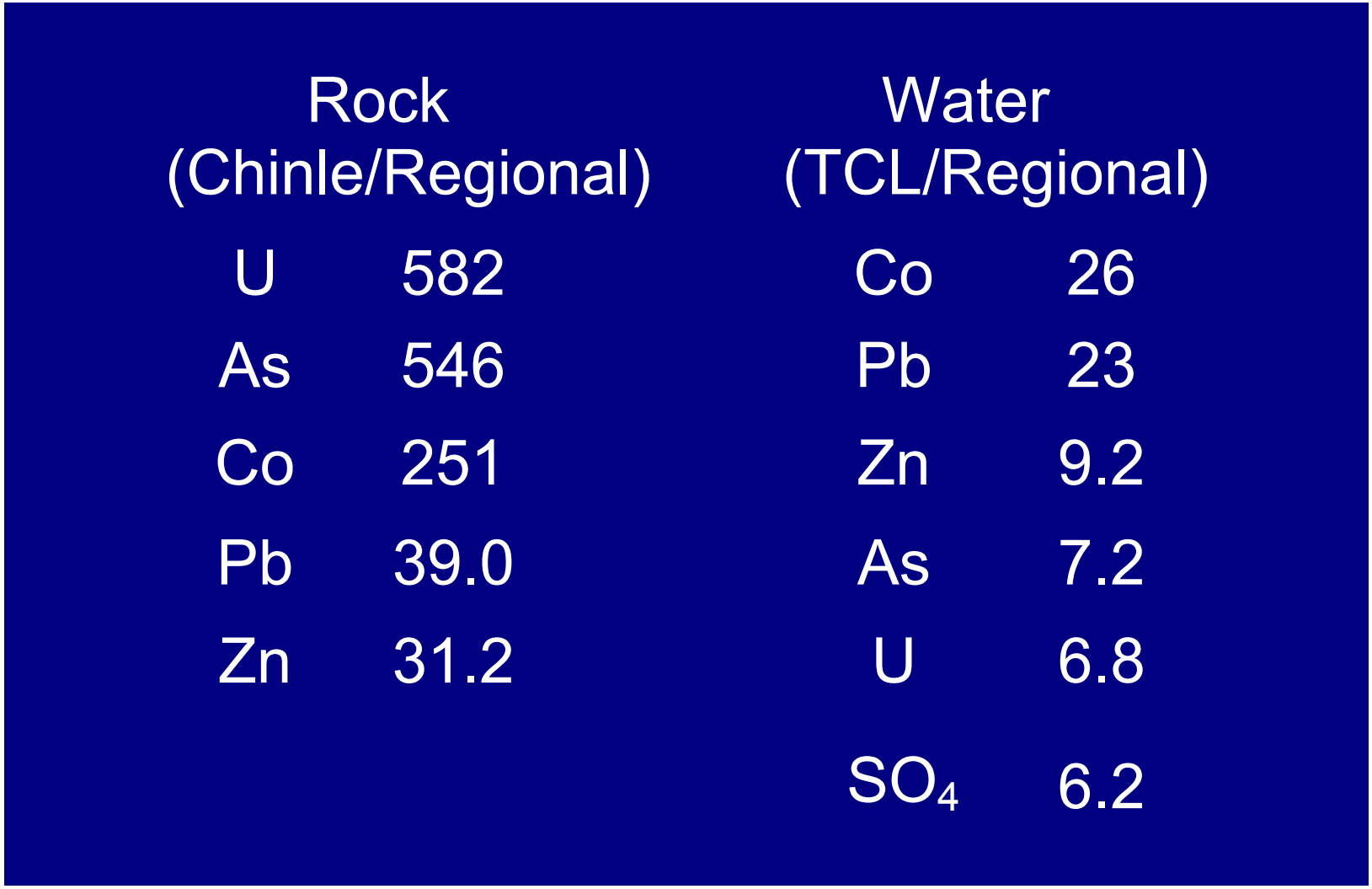

Figure 16. Summary of the elements with the top five greatest ratios of median values in decreasing order. For rock samples, Chinle Formation results are divided by the regional rock data, and for the water samples the Tuba City Landfill results are divided by the regional water samples.

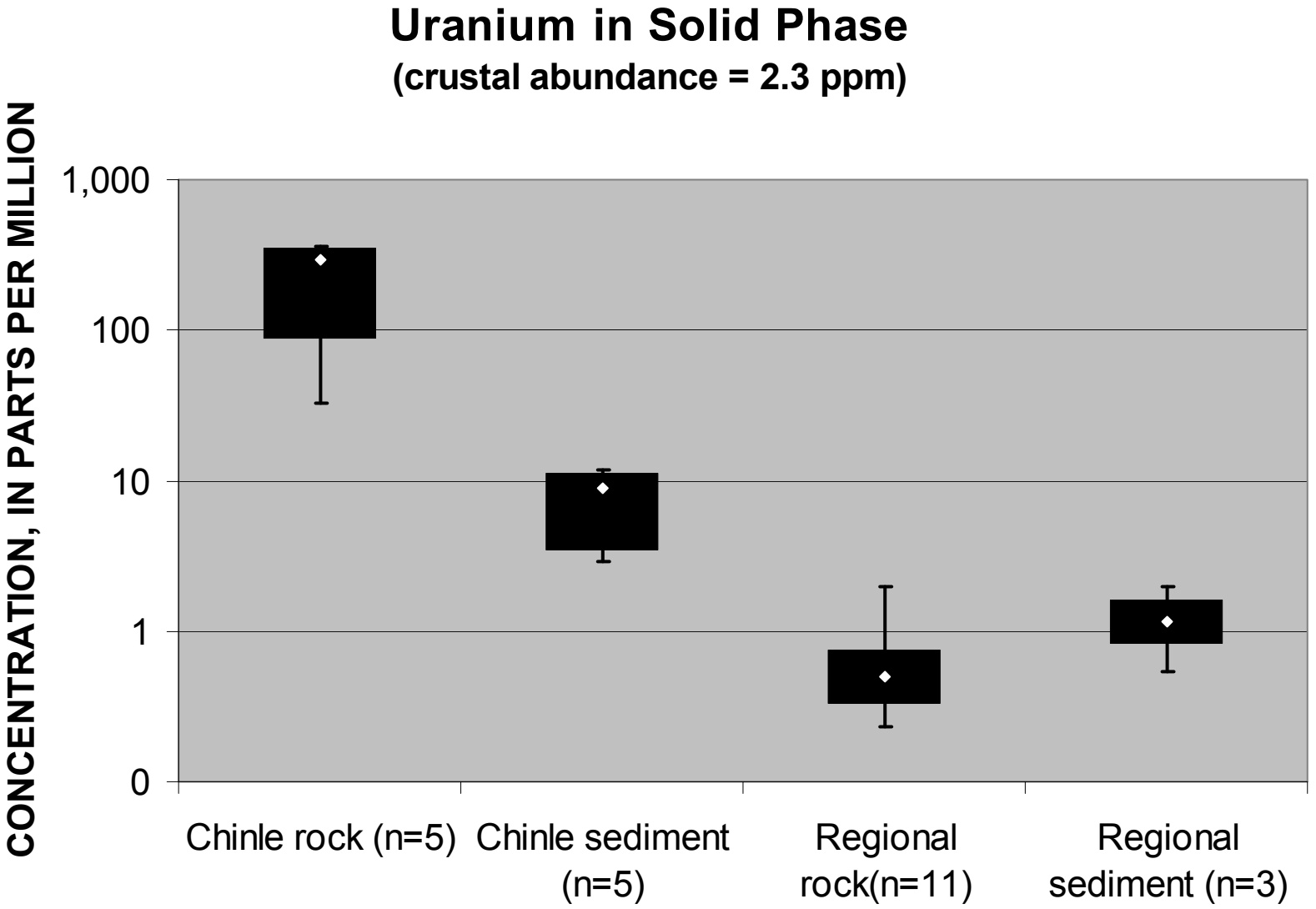

Figure 17. Solid-phase uranium concentrations for the indicated groups of samples in parts per million. "Error bars" indicate the 5th and 95th percentiles. Solid black area indicates the 25th and 75th percentile, and the white diamond indicates the median value. Note the log scale for concentrations. The number of samples is indicated (n).

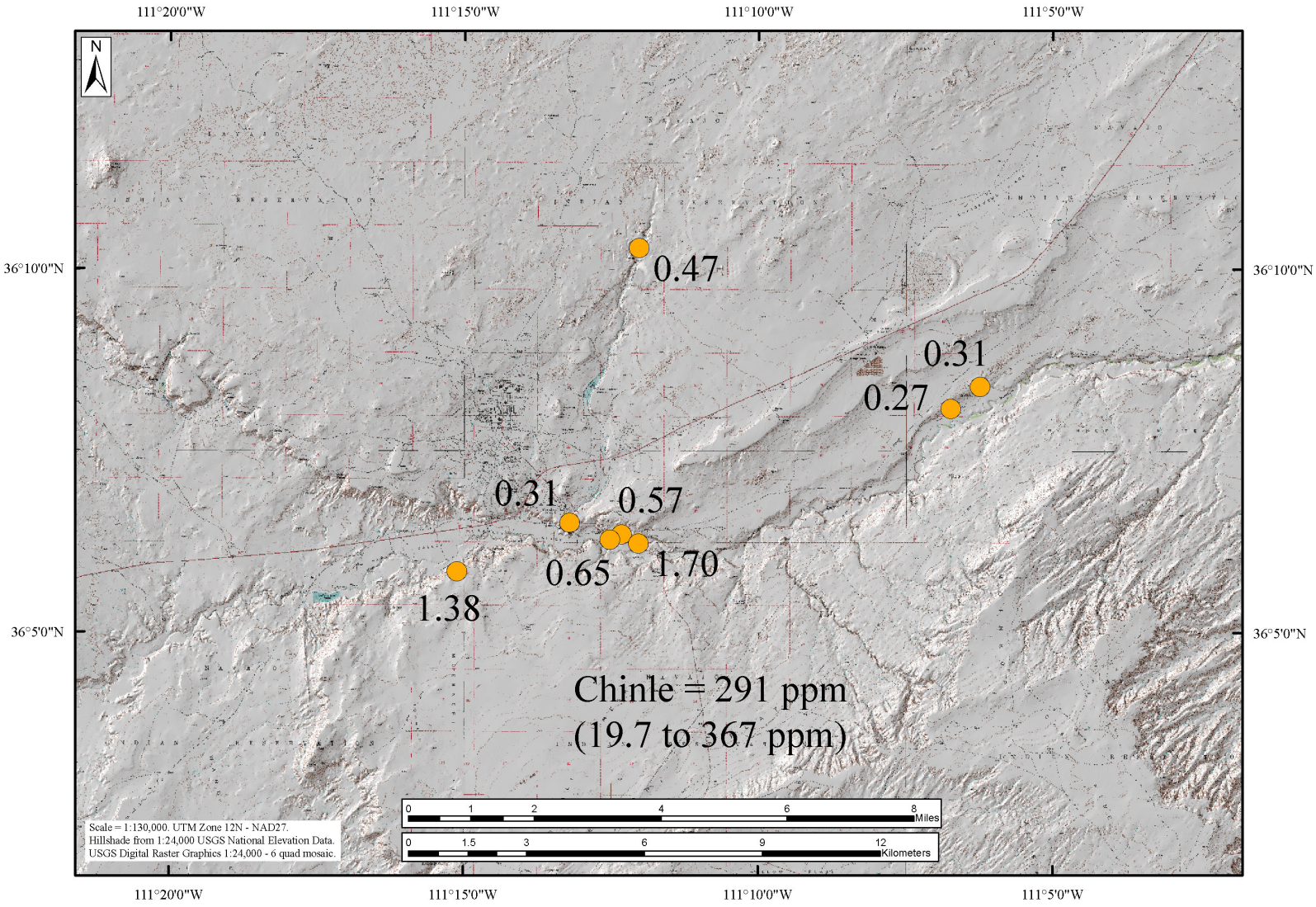

Figure 18. Solid-phase uranium concentrations for regional rock and sediment samples in parts per million. Where multiple samples were taken at the same location, the average value is plotted. Data for the Chinle rock samples are shown for comparison, where listed value is the median and the full range of values is indicated.

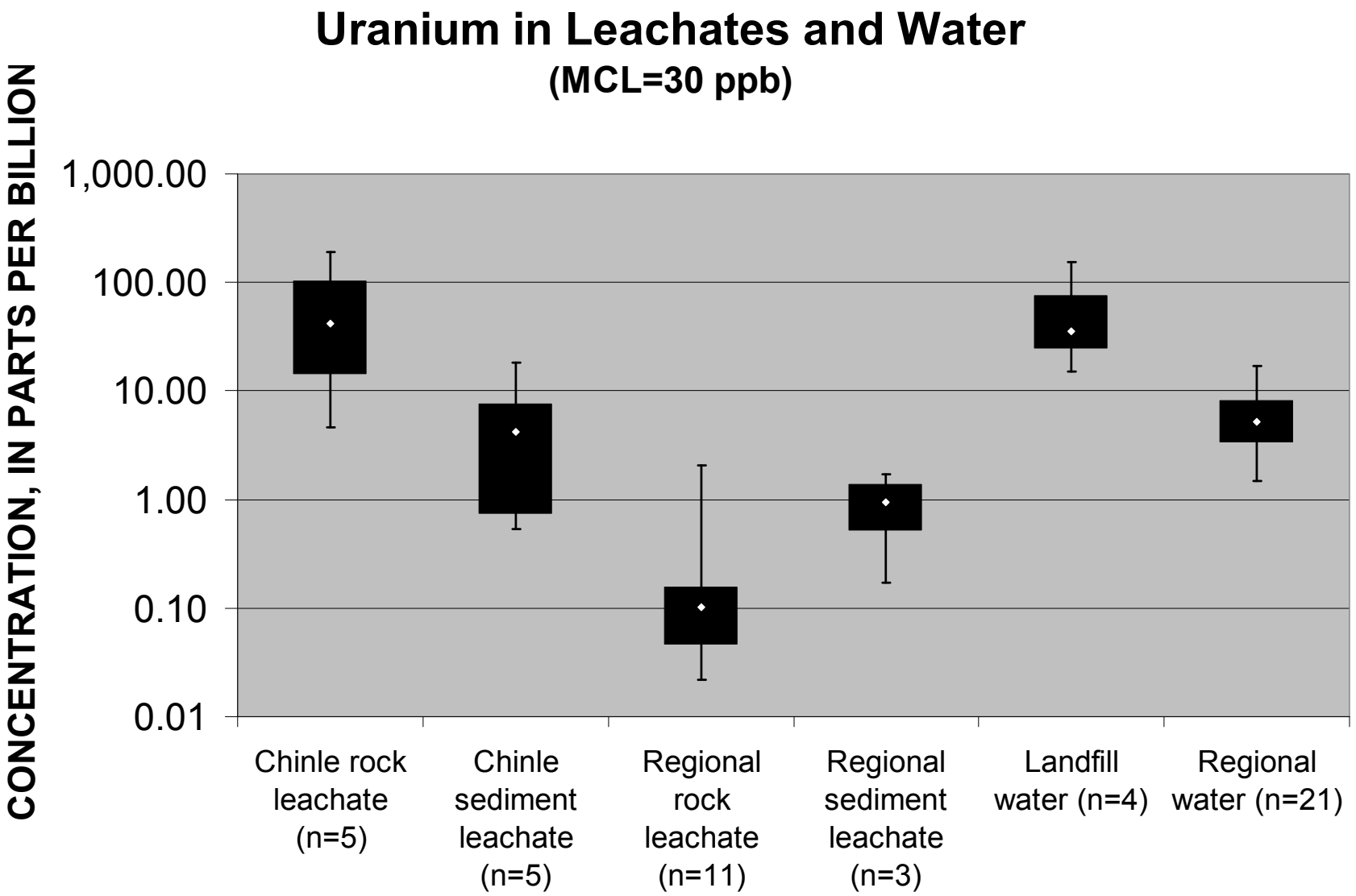

Figure 19. Uranium concentrations for the solid-phase leachates compared to water samples in parts per billion. "Error bars" indicate the 5th and 95th percentiles. Solid black area indicates the 25th and 75th percentile and the white diamond indicates the median value. Note the log scale for concentration. The number of samples is indicated (n).

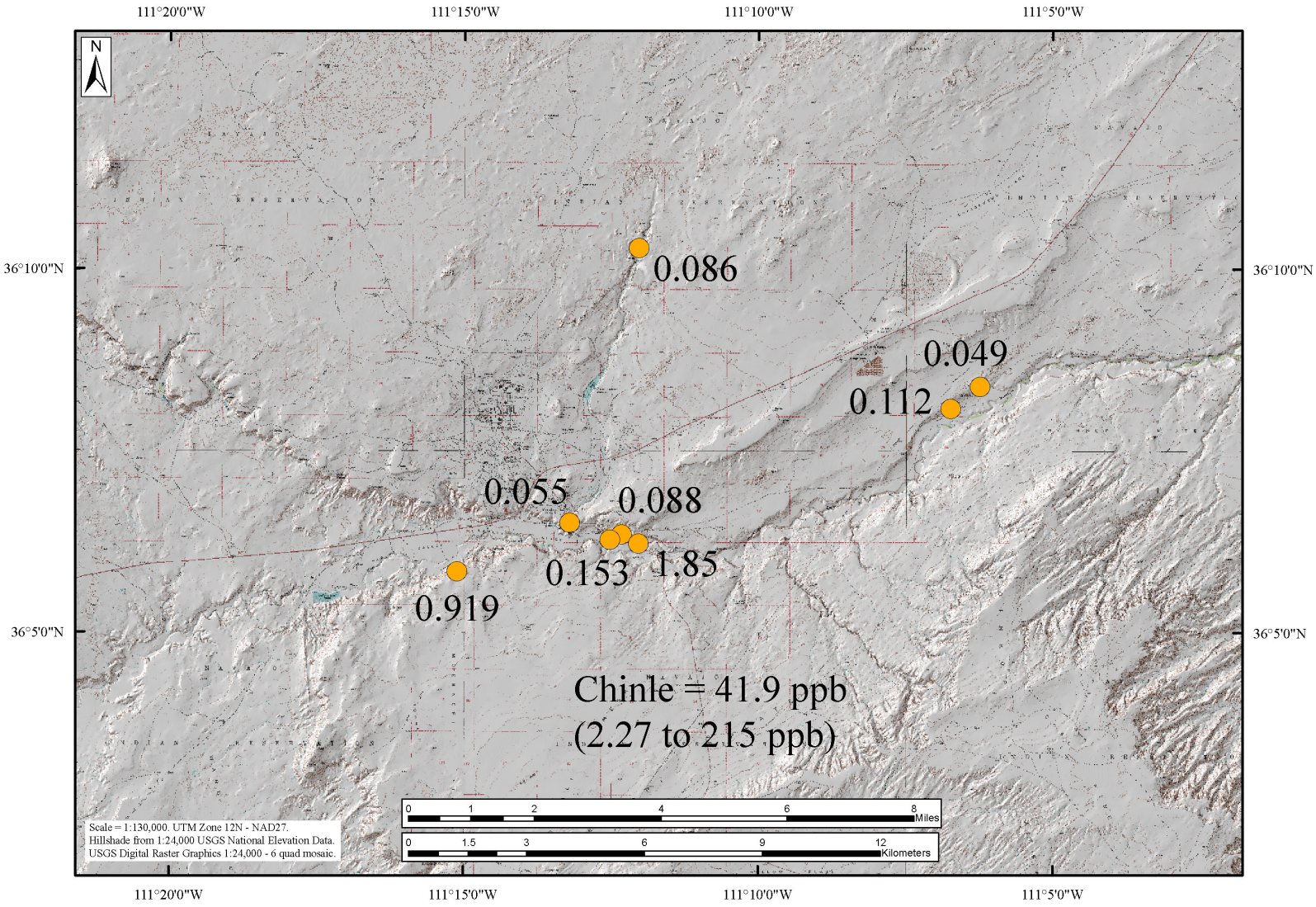

Figure 20. Uranium concentrations for regional rock and sediment-sample leachates in part per billion. Where multiple samples were taken at the same location, the average value is plotted. Data for the Chinle rock samples are shown for comparison, where listed value is the median and the full range of values is indicated.

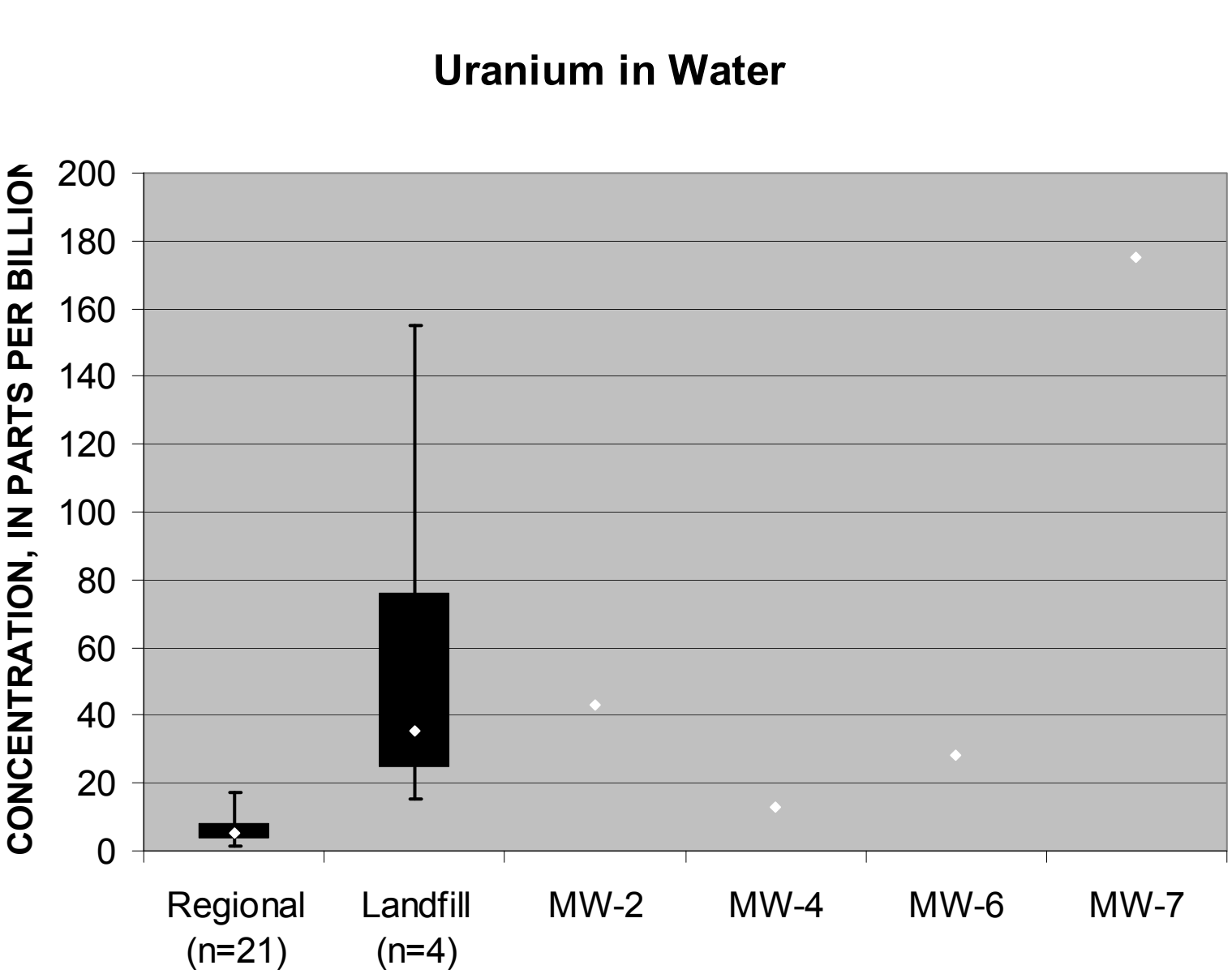

Figure 21. Uranium concentrations in regional and landfill waters with individual landfill wells listed (all in parts per billion). "Error bars" indicate the 5th and 95th percentiles. Solid black area indicates the 25th and 75th percentile, and the white diamond indicates the median value. The number of samples is indicated (n).

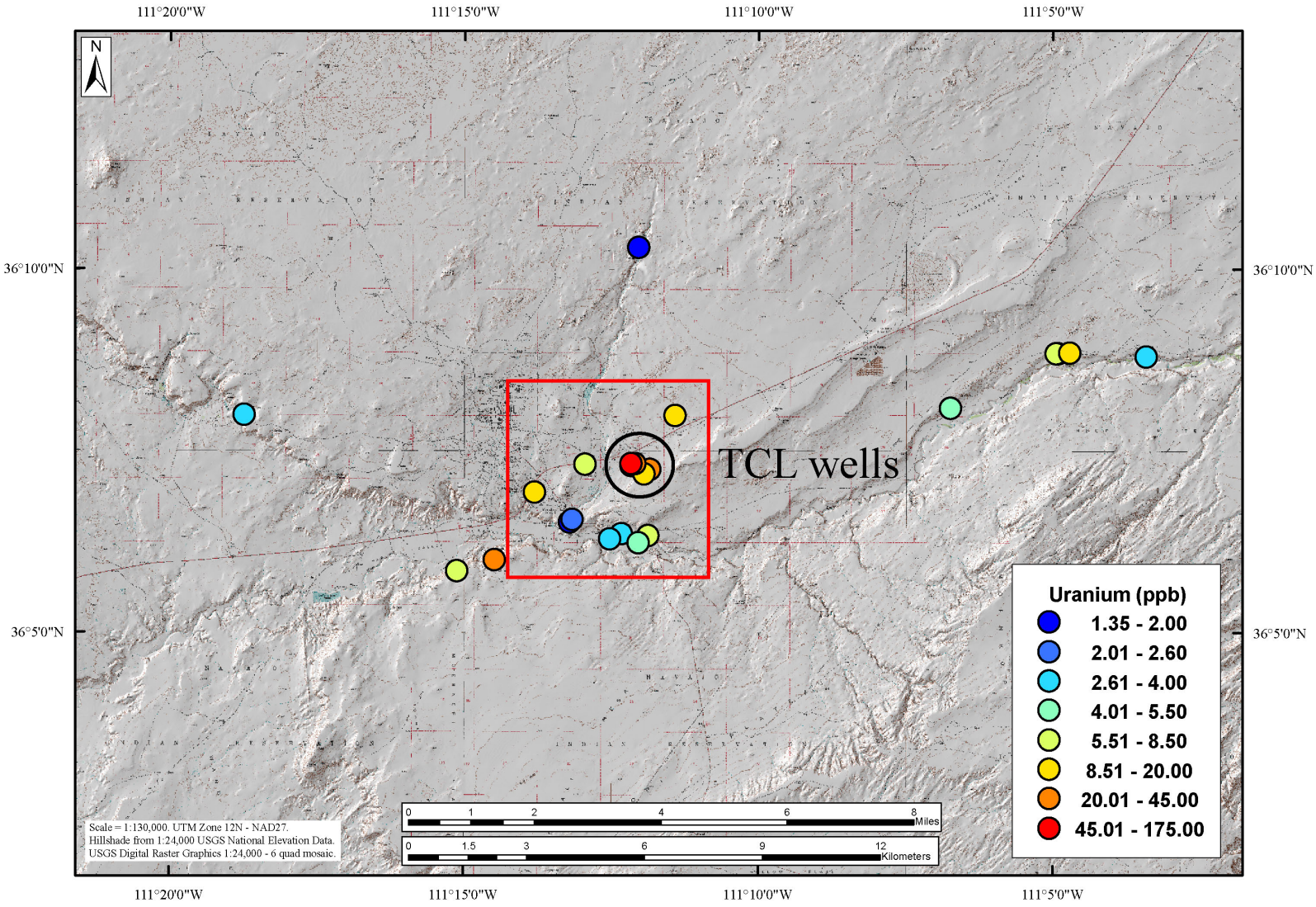

Figure 22. Map of uranium concentrations in water samples in parts per billion. Concentration values are color coded as indicated. Tuba City Landfill wells are circled. Red box indicates enlarged area in next figure.

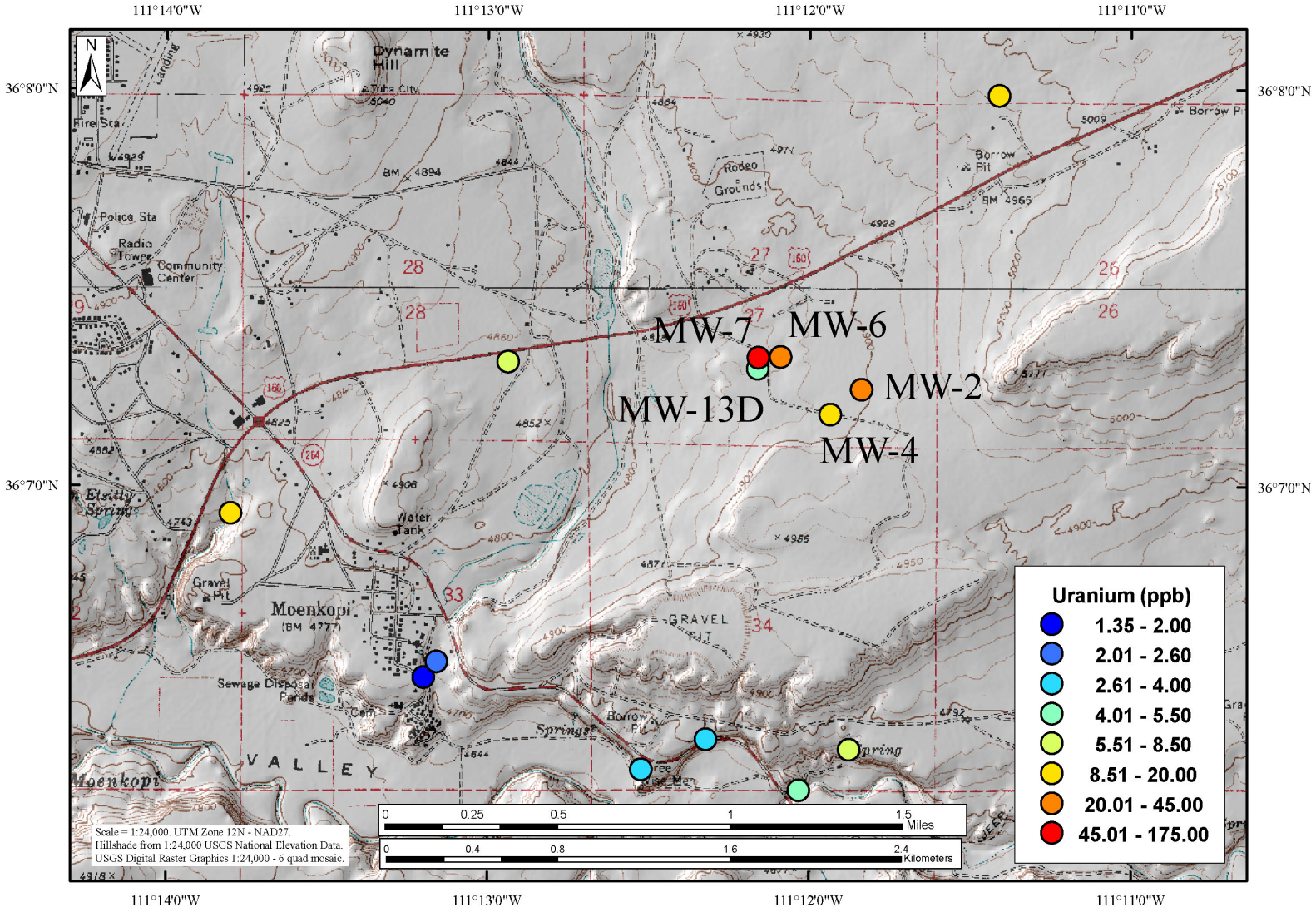

Figure 23. Enlarged area from figure 22 for uranium concentrations in water samples in parts per billion. Concentration values are color coded as indicated. Tuba City Landfill wells are labeled appropriately.

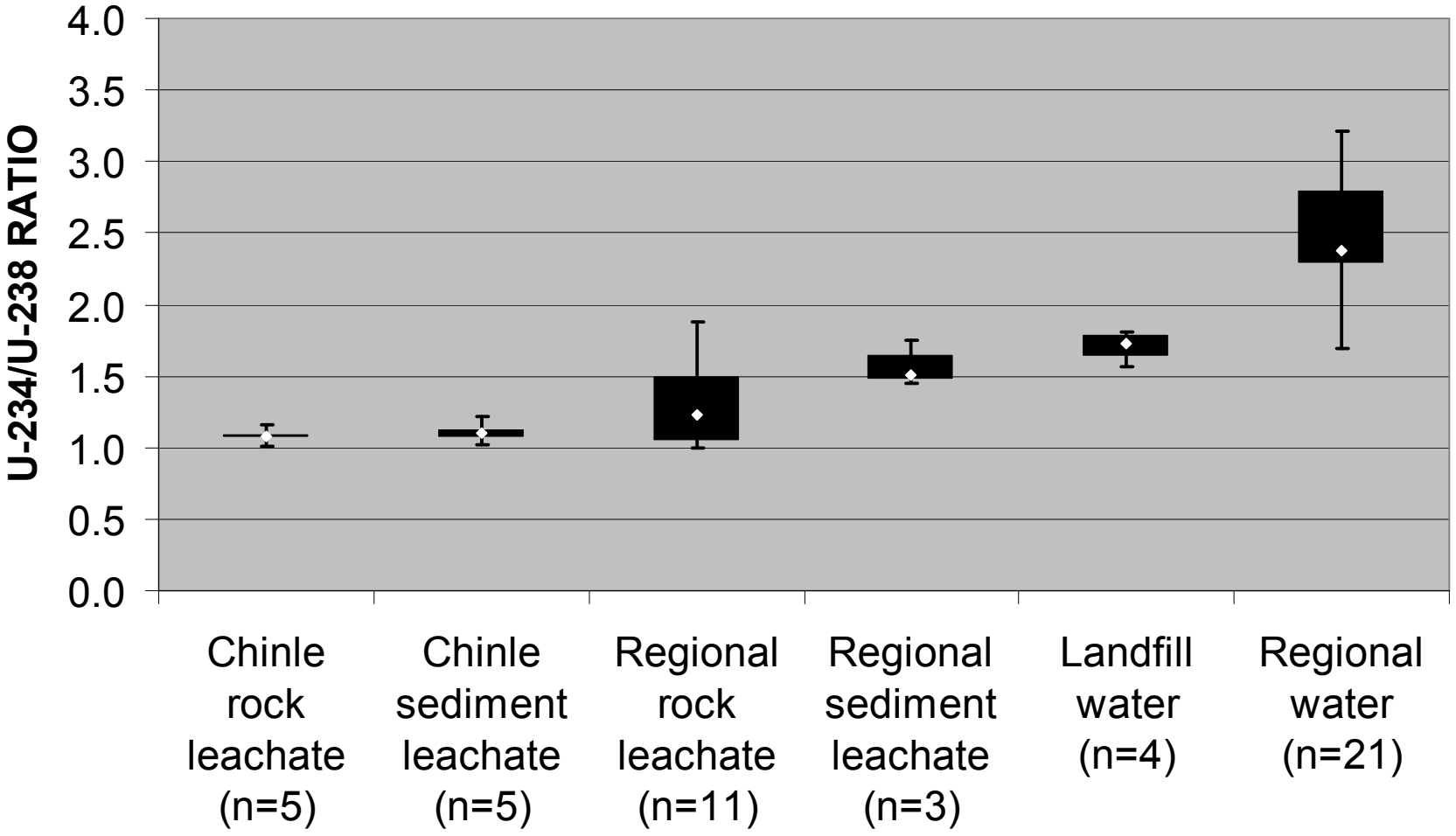

Figure 24. Ratio of $^{234}U/^{238}U$ in rock and sediment leachates compared to landfill and regional waters. "Error bars" indicate the 5th and 95th percentiles. Solid black area indicates the 25th and 75th percentile, and the white diamond indicates the median value. The number of samples is indicated (n).

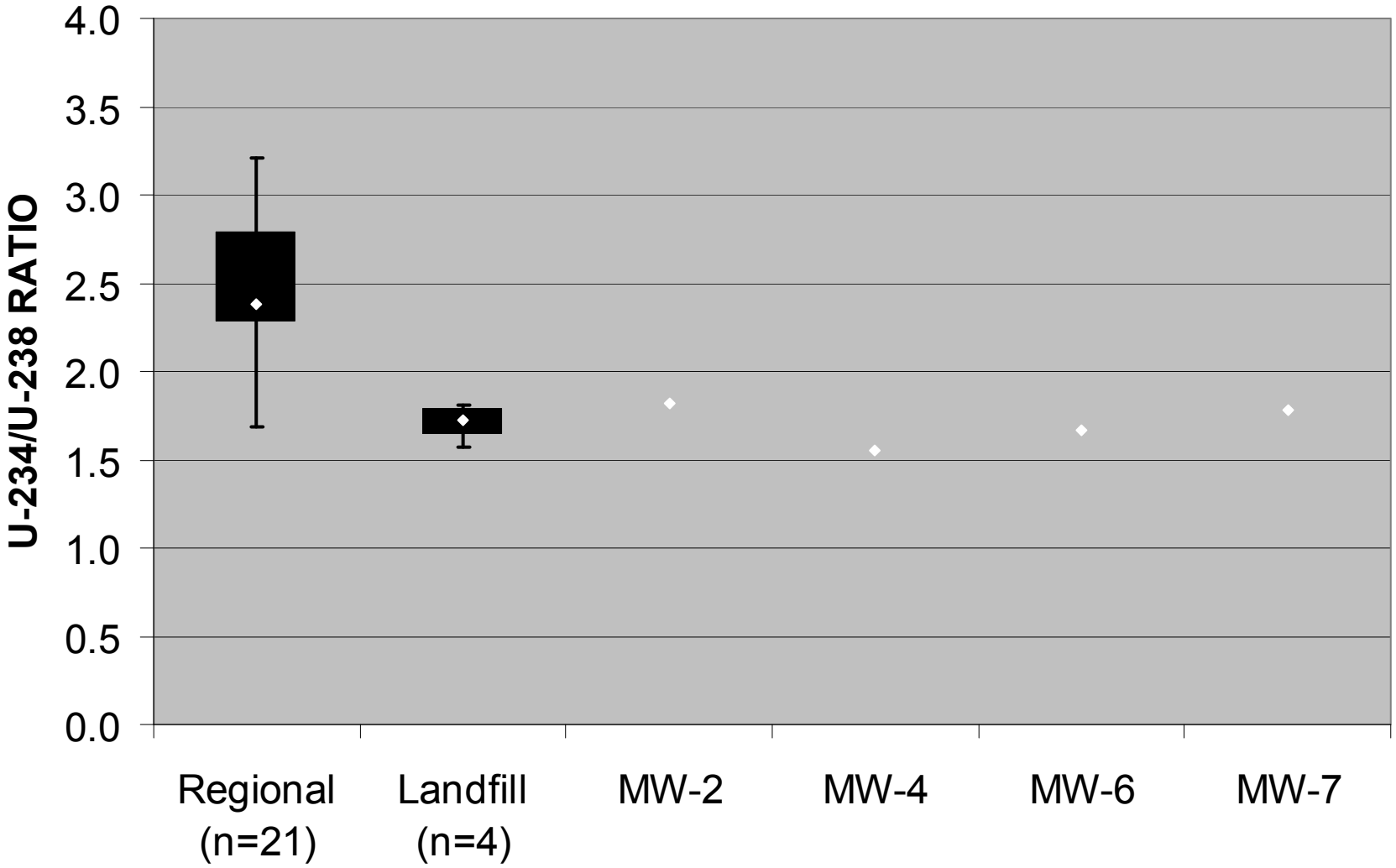

Figure 25. Ratio of $^{234}U/^{238}U$ in regional and landfill waters with individual landfill wells listed. "Error bars" indicate the 5th and 95th percentiles. Solid black area indicates the 25th and 75th percentile, and the white diamond indicates the median value.

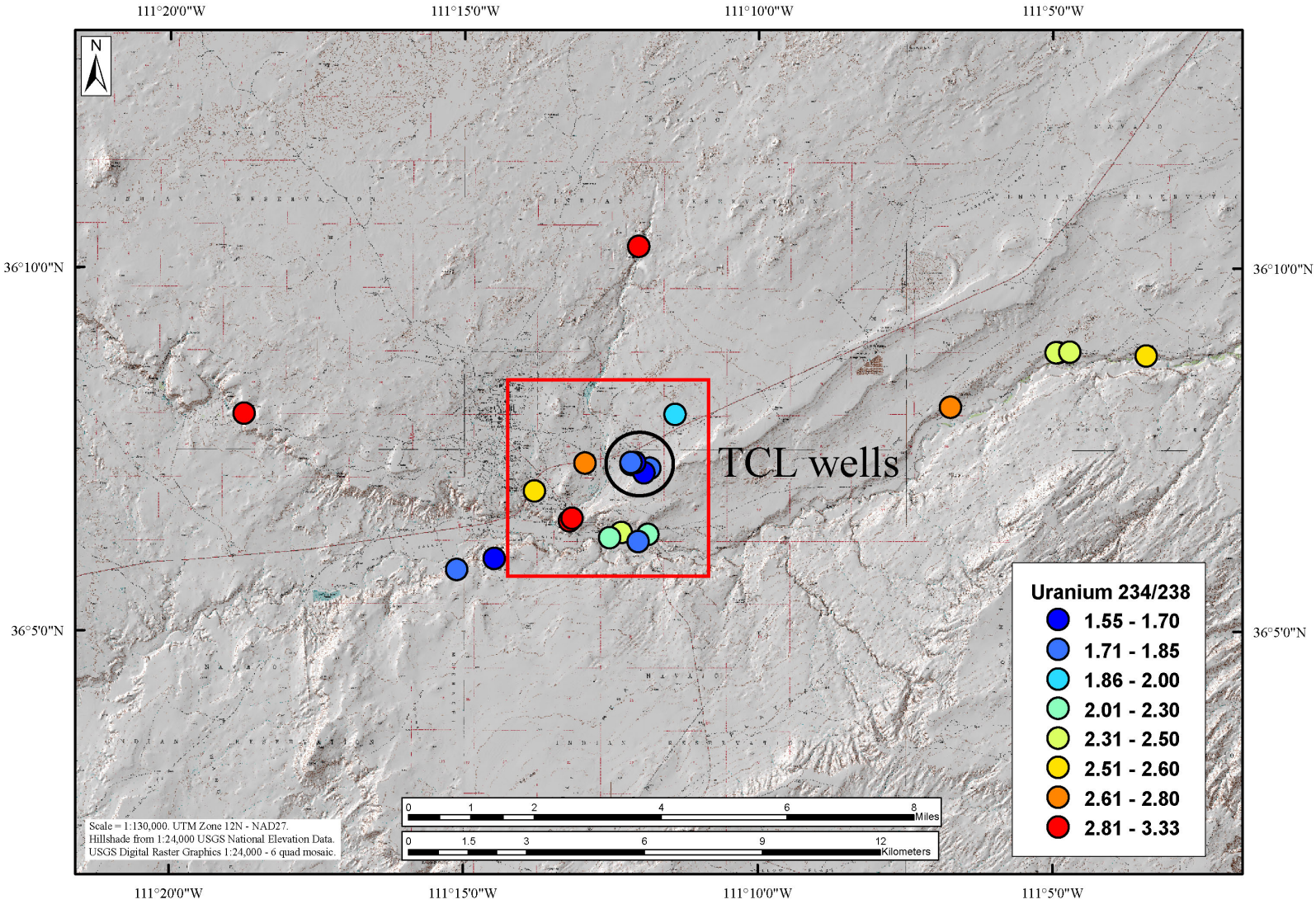

Figure 26. Map of the ratio of $^{234}U/^{238}U$. Ratio values are color coded as indicated. Tuba City Landfill wells are circled. Red box indicates enlarged area in next figure. Lower ratios (blue) indicate shorter rock/water interaction times, and higher ratios (red) indicate longer rock/water interaction times.

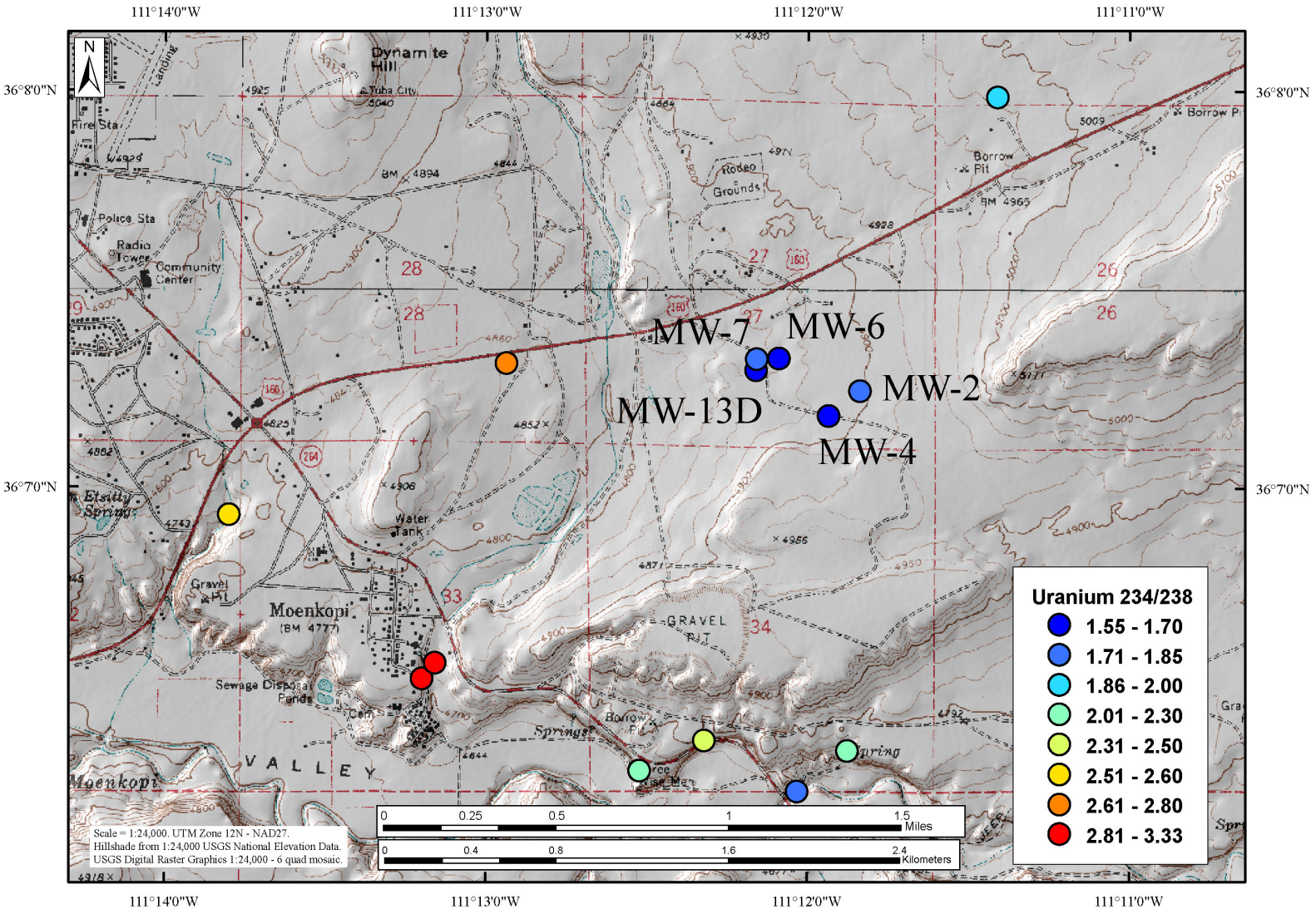

Figure 27. Enlarged area from figure 26 for uranium isotope ratios. Ratio values are color coded as indicated. Tuba City Landfill wells are labeled appropriately. Lower ratios (blue) indicate shorter rock/water interaction times, and higher ratios (red) indicate longer rock/water interaction times.

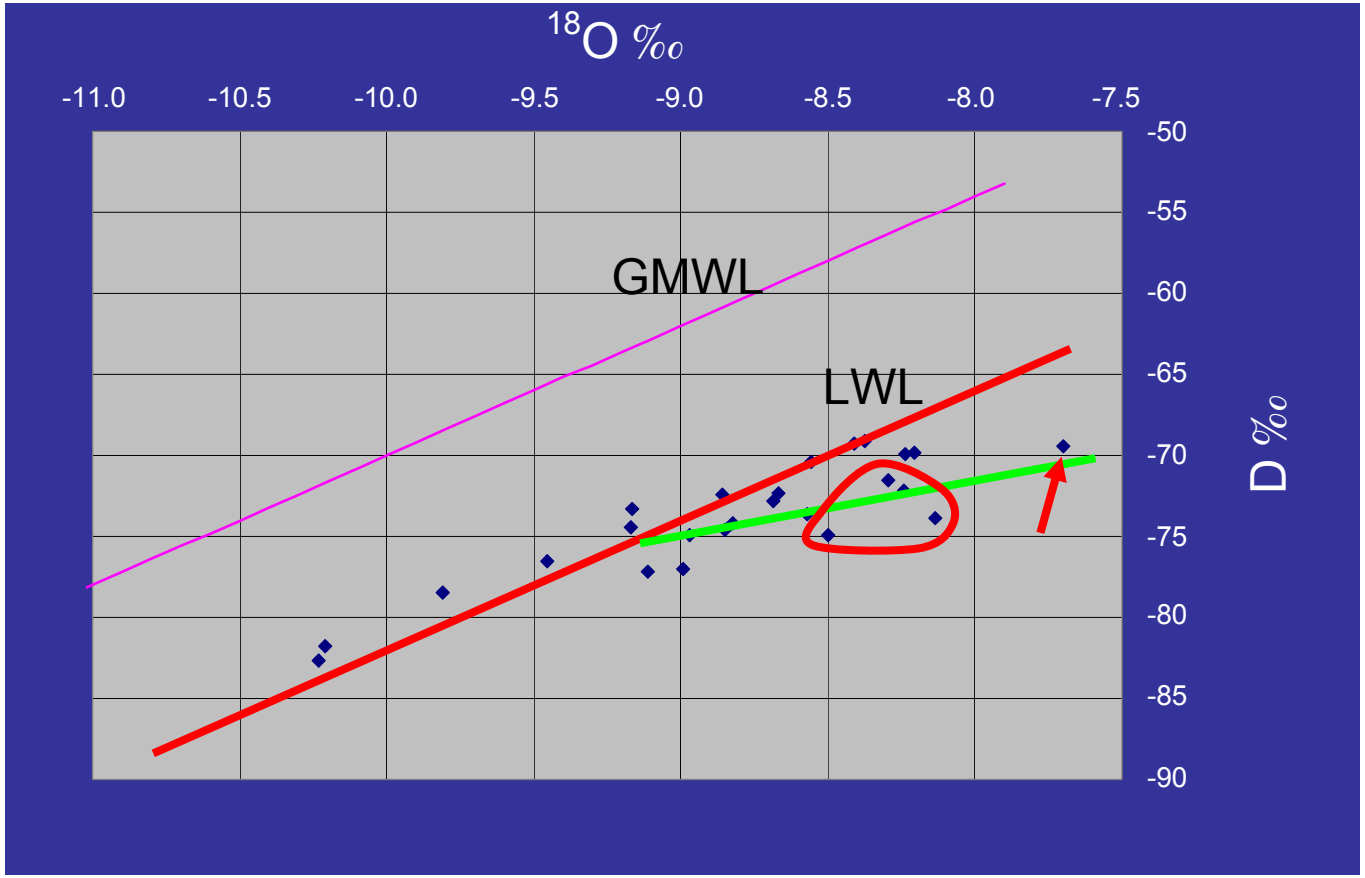

Figure 28. Oxygen (δ ^{18}O)and deuterium (δ D) isotopes for regional and landfill waters. GMWL = global meteoric water line and LWL = local water line. Landfill wells (MW-2, 4, 6, and 7) are circled, and arrow points to the Herbert Chee well (06TCSP107), a hand-dug well just upgradient from the TCL. Green line indicates the approximate evaporative line that is a smaller slope than the LWL.

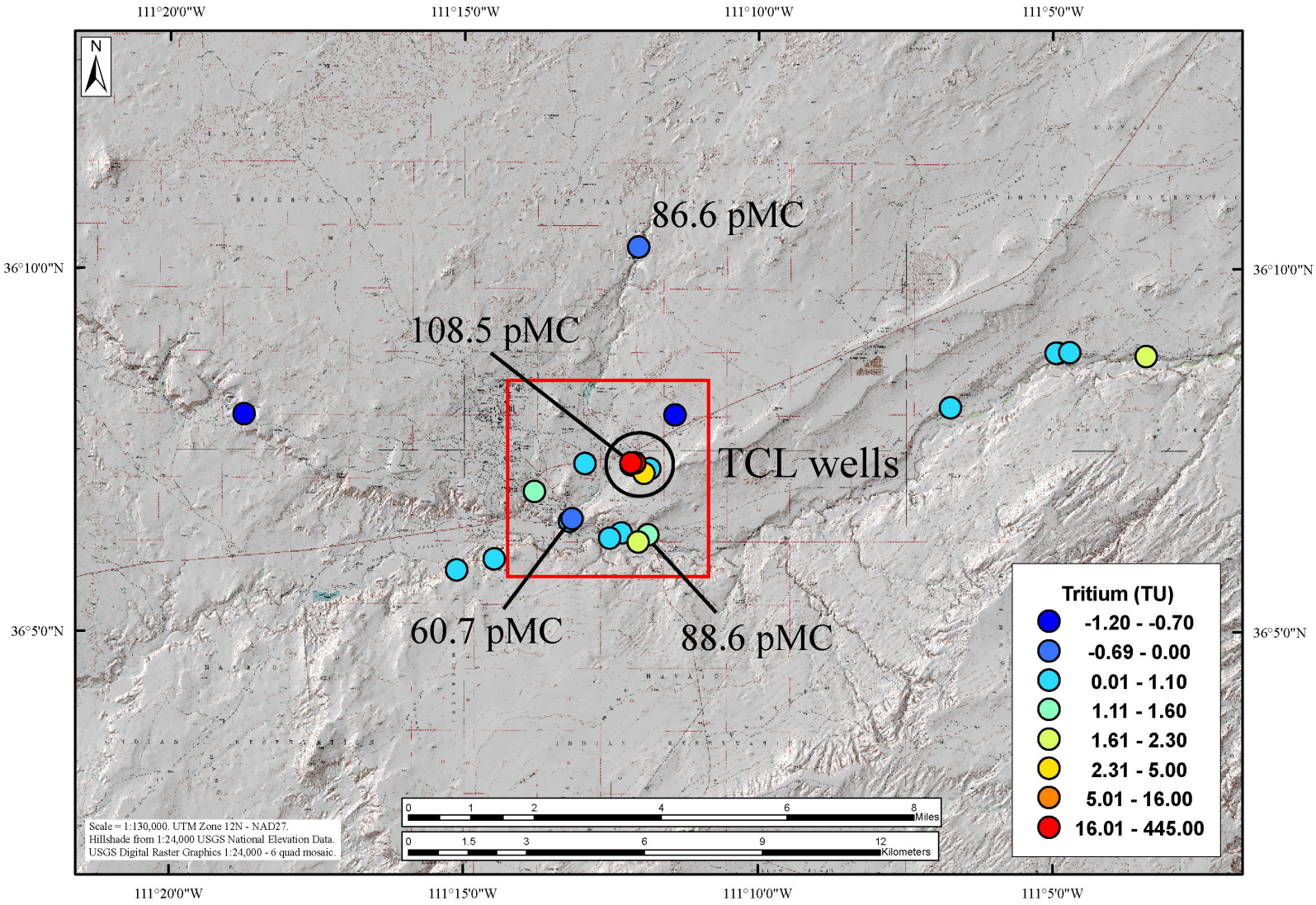

Figure 29. Concentrations of tritium (in tritium units, TU) and ^{14}C (in pMC, percent modern carbon) in the regional and landfill waters. Tritium values are color coded as indicated. ^{14}C concentrations are labeled directly. Tuba City Landfill wells are circled. Red box indicates enlarged area in next figure. Negative values are below detection limits. Low tritium values (less than 0.8 TU) indicate ages greater than 60 years, and higher tritium values (more than 0.8 TU) indicate a portion of the water is less than 60 years old.

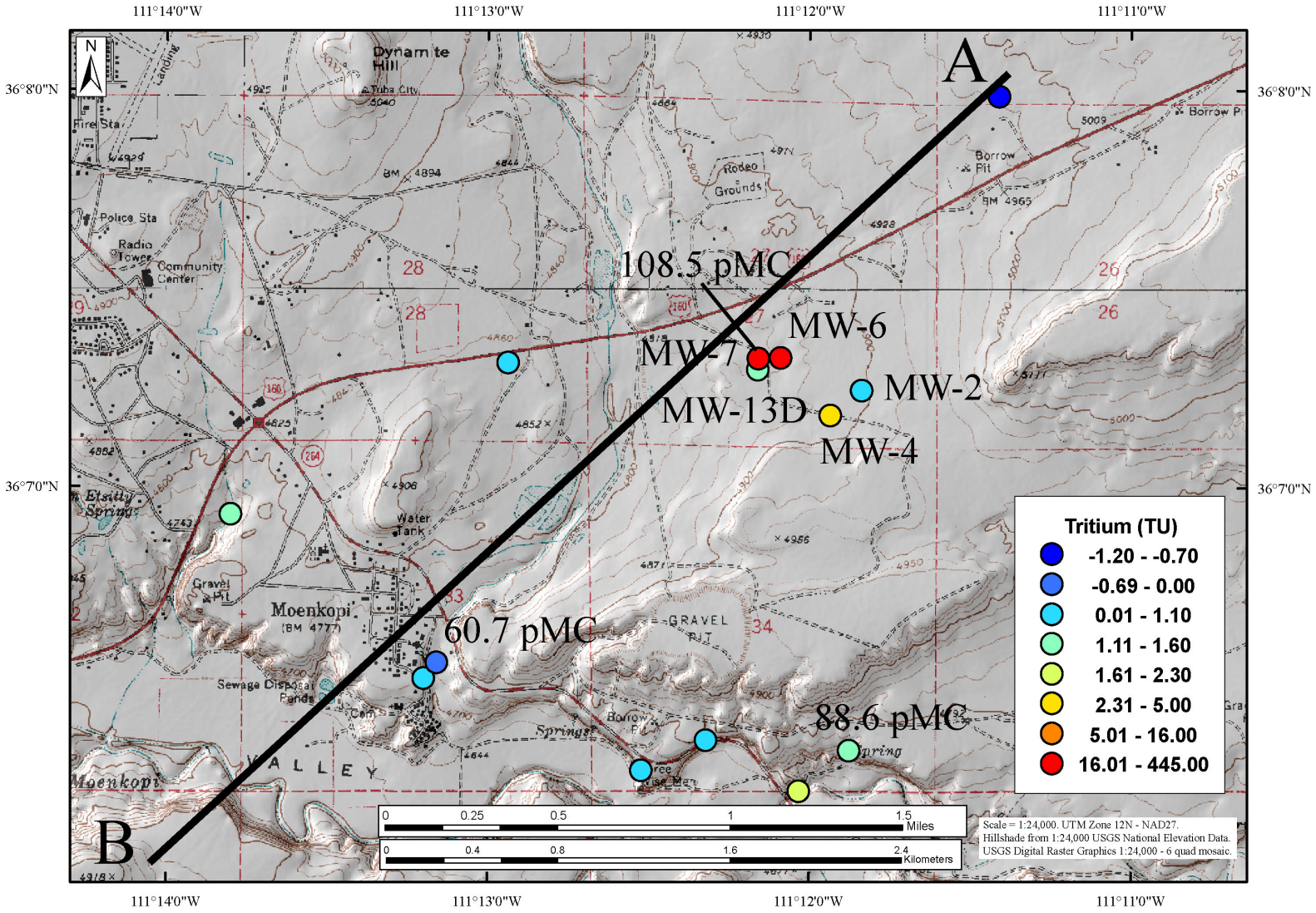

Figure 30. Enlarged area from figure 29 for tritium (in tritium units, TU) and ^{14}C (in pMC, percent modern carbon) concentrations. Tritium values are color coded as indicated. 14C concentrations are labeled directly. Tuba City Landfill wells are labeled appropriately. Line A–B is the location of a cross section presented in the next figure. Negative values are below detection limits. Low tritium values (less than 0.8 TU) indicate ages greater than 60 years, and higher tritium values (more than 0.8 TU) indicate a portion of the water is less than 60 years old

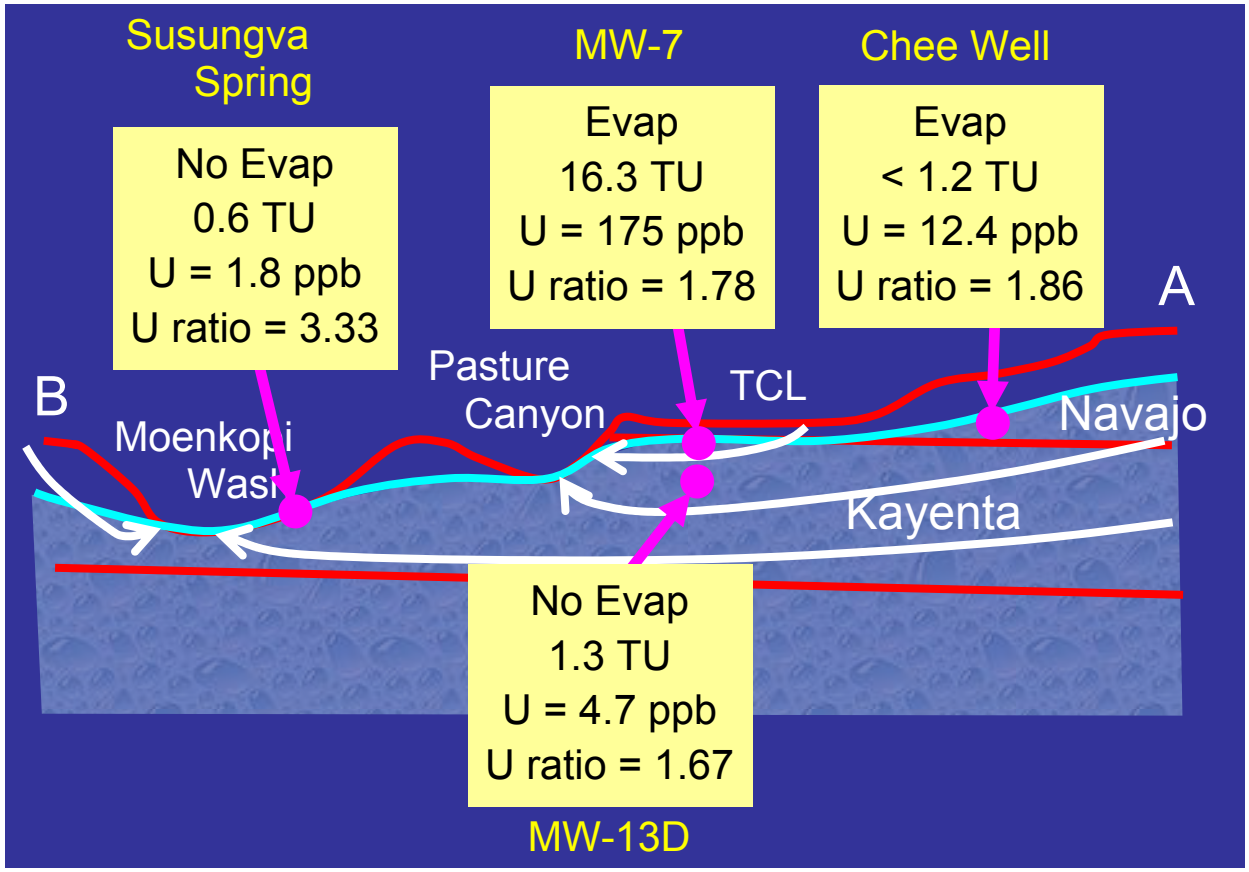

Figure 31. Conceptual cross section for line A–B in figure 30. Evaporation, tritium concentration, uranium concentration, and uranium isotope ratios are indicated to highlight the ground-water flow paths. TU, tritium units; ppb, parts per billion.

Table 1. Analyses completed.

[ICP, ion coupled plasma; MS, mass spectrometry; IC, ion chromatography; AES, atomic emission spectrometry; U, uranium, Sr, strontium; S, sulfur]

Number of samples	Laboratory	USGS, Denver[1]	USGS, Denver[1]	USGS, Denver[1]	USGS, Denver[2]	UA, Tucson[3]	UA, Tucson[3]	NAU, Flagstaff[4]	CU Boulder[5]	UA, Tucson[3]
	Method	**ICP–AES**	**ICP–MS**	**IC**	**Isotope ratio MS**	**Liquid scintillation counting**	**Isotope ratio MS**	**Sector field ICP–MS**	**Isotope ratio MS**	**Isotope ratio MS**
	Type of sample	**27-Element cations**	**40-Element cations**	**Major anions**	**Stable isotopes of oxygen and hydrogen**	**Tritium**	**Carbon isotopes**	**$^{234}U/^{238}U$ Activity ratio**	**^{87}Sr**	**^{34}S**
25	Surface and ground water	x	x	x	x	x	x	x		x
24	Solid-phase leachates (filtered)	x	x				x	x	x	selected
8	Stream sediment (solid)		x							
16	Rock samples (solid)		x							

[1] USGS Mineral Resources Program (MRP) laboratory, Building 20, Denver Federal Center.

[2] USGS Crustal Imaging and Characterization Team (CICT) stable-isotope laboratory, Building 25, Denver Federal Center.

[3] University of Arizona, Tucson, Department of Geosciences Laboratory of Isotope Geochemistry, Christopher J. Eastoe.

[4] Northern Arizona University, Flagstaff, Chemistry Department, Dr. Michael Ketterer.
(analysis for U-activity ratio and U-concentration by MS on 50 mL)

[5] University of Colorado at Boulder, Department of Geosciences, Lang Farmer.