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Assessing field-scale migration of mobile radionuclides at the Nevada Test Site

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Abstract

Numerous long-lived radionuclides, including ^{99}Tc (technetium) and ^{129}I (iodine), are present in groundwater at the Nevada Test Site (NTS) as a result of 828 underground nuclear weapons tests conducted between 1951 and 1992. We synthesize a body of groundwater data collected on the distribution of a number of radionuclides (^3H , ^{14}C , ^{36}Cl , ^{99}Tc and ^{129}I), which are presumably mobile in the subsurface and potentially toxic to down-gradient receptors, to assess their migration at NTS, at field scales over distances of hundreds of meters and for durations of more than thirty years. Qualitative evaluation of field-scale migration of these radionuclides in the saturated zone provides an independent approach to validating their presumably conservative transport in the performance assessment of the proposed geological repository at Yucca Mountain, which is located on the western edge of NTS. The analyses show that the interaction of ^3H with a solid surface via an isotopic exchange with clay lattice hydroxyls may cause a slight delay in the transport of ^3H . The transport of ^{14}C could be retarded by its isotopic exchange with carbonate minerals, and the exchange may be more pronounced in the alluvial aquifer. In particular, ^{99}Tc may not necessarily exist as a mobile and conservative species $^{99}\text{TcO}_4^-$, as commonly assumed for NTS groundwater. This is corroborated with recent *in situ* redox potential measurements, both across and near Yucca Mountain, showing that groundwater at multiple locations is not oxidizing. Speciation of iodine and its associated reactivity and mobility is also complex in the groundwater at the NTS and deserves further attention. The assumption of no retardation for the transport of ^{99}Tc (especially) and ^{129}I , used at the performance assessment of Yucca Mountain repository, is probably overly conservative and results in unrealistically high estimated doses for down-gradient receptors.

23

Keywords: radionuclide, transport, field-scale, saturated zone, Yucca Mountain, rock.

1 1. Introduction

2 In studies of radionuclide transport and retardation at Yucca Mountain, the sorption
3 distribution coefficients (K_d) approach has been employed to quantify the extent of radionuclide-
4 groundwater-rock interactions. Values of K_d are relied upon in transport codes to take into
5 account the delayed transport of sorbing radionuclides from retardation. Unrepresentative K_d
6 values will lead to unrealistic predictions of radionuclide transport. The K_d data, used in the Total
7 System Performance Assessment (TSPA) analyses for the proposed Yucca Mountain repository,
8 are based on laboratory batch (mostly) and column experiments, supplemented with literature
9 information (CRWMS M&O, 2000). For the radionuclides important to TSPA, carbon and
10 chlorine are deemed to be nonsorbing (with K_d value of 0) in all representative (devitrified,
11 vitric, and zeolitized) tuffs encountered below the potential repository horizon. The
12 recommended TSPA values of K_d for ^{99}Tc and ^{129}I in the saturated zone are zero (nonsorbing) in
13 all volcanic tuffs and between 0.27 and 0.63 mL/g (weakly sorbing) in alluvium (CRWMS
14 M&O, 2000). These values are based on the assumption that Tc and I exist as pertechnetate
15 (TcO_4^-) and I^- in YMP groundwaters and on experimental measurements of K_d values under
16 oxidizing conditions.

17 However, both Tc and I are redox-sensitive radionuclides, and the speciation and
18 retardation of these radionuclides are sensitive to modest changes in redox potential potentially
19 occurring in the groundwater flow pathways. With the probable presence of groundwaters with
20 variable redox conditions along the potential transport pathways, these radionuclides could have
21 much longer transport times than those predicted by the current TSPA calculations. Valuable
22 field-scale information to elucidate the natural controls on the mobility of these species could be
23 garnered from the groundwater migration experiments and monitoring program that has taken

1 place over the past thirty years to assess contamination resulting from the underground nuclear
2 tests conducted at the NTS.

3 As discussed by Smith et al. (2001), NTS provides an analog to assess the performance of
4 Yucca Mountain nuclear waste repository in the absence of engineered barriers (i.e.,
5 performance from natural barriers system). At the NTS, a large inventory of radionuclides
6 (tritium, fission products, activation products, and actinides) is present at various concentrations
7 as a result of the 828 underground nuclear weapons tests. Since 1973, various programs have
8 investigated the environmental effects of nuclear testing at the NTS. An appreciable body of
9 information regarding radionuclide distribution at NTS has been accumulated, and a synthesis of
10 this information provides insight into field-scale radionuclide migration. Because of their
11 comparable hydrogeologic settings, understanding gained from NTS is directly applicable to the
12 performance of the proposed Yucca Mountain repository, which is located on the western edge
13 of NTS (Figure 1).

14 A similar suite of radionuclides present in the waste to be buried at the potential Yucca
15 Mountain repository that are important for TSPA analyses are also present in the groundwater at
16 NTS from the underground nuclear testing. Among these radionuclides are ^3H (tritium), ^{14}C
17 (carbon), ^{36}Cl (chlorine), ^{99}Tc (technetium), and ^{129}I (iodine) that presumably have high potential
18 for migration, as well as sorptive ^{90}Sr (strontium), ^{137}Cs (cesium), and uranium and plutonium
19 isotopes. Most nuclear tests at NTS were conducted between ~500 and 1200 m below the land
20 surface. Approximately 300 nuclear tests were detonated either below or within 100 m of the
21 water table; the cavities produced by these tests have since refilled with water and subsequent
22 radionuclide migration, examined in various monitoring wells in different geologic media, in the
23 saturated zone forms the basis of this work. The geologic media that hosted the underground
24 tests at NTS consists primarily of tuffs, rhyolites, and tuffaceous alluvium, which are very

1 similar to the various geological media encountered in the TSPA analyses of the potential
2 repository at Yucca Mountain. Yucca Mountain consists of a group of north–south-trending
3 block-faulted ridges composed of tertiary volcanic rocks. At approximately 12–13 kilometers
4 from the Mountain, the water table transitions from volcanic tuffs to alluvium (Eddebarh et al.,
5 2003).

6 By synthesizing available NTS data, a further understanding of the migration behavior of
7 these radionuclides at NTS can be obtained, in a cost-effective manner, at field scales over
8 distances of hundreds of meters and for durations of more than 30 years and under hydrogeologic
9 conditions similar to the proposed Yucca Mountain repository. The behavior and magnitude of
10 sorption for these radionuclides used in TSPA analyses for Yucca Mountain repository can be
11 validated by comparing NTS observations over a large scale both in space and time. With their
12 high activity in the waste, long half-life (2.13×10^5 and 1.57×10^7 for ^{99}Tc and ^{129}I , respectively),
13 and presumed conservative transport behavior, both ^{99}Tc and ^{129}I are the important dose
14 contributors to the calculated health risk. For example, in the transport simulations in the
15 unsaturated zone below the potential repository, 10% and 50% of released nonsorbing ^{99}Tc will
16 reach the water table (~ 300 m below the repository horizon) at 300 and 4,000 years, respectively
17 (Moridis et al., 2000). After arriving in the saturated zone, the transport times for nonsorbing
18 radionuclides (such as ^{129}I and ^{99}Tc) are on the order of hundreds of years to reach the
19 compliance boundary located 18 km from the potential repository (Arnold et al., 2003;
20 Eddebarh et al., 2003). A slight sorption in rock matrix will significantly reduce the transport
21 times. For example, 10% and 50% of released ^{237}Np , with K_d values 1 mg/L for devitrified and
22 vitric tuff and 4 mg/L for zeolitized tuff, will travel through the unsaturated zone as long as
23 10,000 and 120,000 years, respectively (Moridis et al., 2000). The understanding of field-scale
24 migration behavior of radionuclides, such as ^{99}Tc , obtained from the NTS data can be used to

1 examine their transport behavior, and reduce the conservatism in input parameters adopted in the
2 TSPA analyses.

3 **2. Phenomenology of nuclear testing and radionuclide distribution**

4 We briefly review the phenomenology, which describes the effects of the nuclear
5 explosion on the surrounding geologic medium, of a subsurface nuclear weapons test, because
6 the initial processes determine the radiologic source term that is available for hydrologic
7 transport. The following description is mainly derived from Borg et al. (1976), Smith (1995),
8 IAEA (1998), and Tompson et al. (2002).

9 The initial detonation produces a high temperature (up to 10^8 °C) and pressure (above
10 10^{10} Pa) shock wave that moves outward and vaporizes the emplacement canister and
11 surrounding rock. This forms a cavity with its growth stops within a few tenths of a second when
12 the pressure drops to the weight of the overburden. Geologic materials outside the immediate
13 cavity are then altered through melting, compression, or fracturing. A few seconds after the
14 shock and elastic waves have dissipated, the less-volatile rock vapors begin to condense and
15 gather at the cavity bottom to form a puddle of molten material, which is further condensed into
16 melt glass.

17 The radionuclides deposited in the subsurface as a result of an underground nuclear
18 explosion consist of radionuclides produced from the nuclear reaction (fission products and
19 tritium), neutron activation of the surrounding host medium, and un-reacted nuclear material.
20 The total inventory of residual radioactivity is called the radiologic source term, while the
21 hydrologic source term is the portion of the radiologic source term that is or becomes available
22 for groundwater transport. The partitioning of the radiologic source term among solid, liquid, and
23 gaseous phases are a function of the device design, host geologic media, physic-chemical

1 properties of the specific radionuclides (in particular, volatility), and the rate and character of
2 cavity growth and collapse.

3 Field investigations at NTS have yielded information regarding the initial distribution of
4 radionuclides after a nuclear test, as discussed in Smith (1995). The inferred partitioning of
5 selected radionuclides among glass, rubble, water, and gas is shown in Table 1. The data in Table
6 1 originate from measurements derived from radiochemical diagnostics of nuclear tests (e.g.,
7 Borg et al., 1976), augmented by general thermodynamic properties (e.g., boiling points, vapor
8 pressures) of these elements under extreme conditions. With the exception of ^{90}Sr and ^{137}Cs , the
9 data apply to the general understanding of radionuclide distributions from underground nuclear
10 testing. The partitioning behavior of ^{90}Sr and ^{137}Cs , with their gaseous precursors ^{90}Kr and ^{137}Xe ,
11 between rubble and melt glass is strongly dependent on the cooling time of the glass as well as
12 on the presence of volatile, non-condensable gases (e.g., CO_2 , H_2) in the cavity–chimney region.

13 In general, refractory elements (e.g., Pu, Am, Np, Ce, Eu) with higher boiling points and
14 lower vapor pressures are largely incorporated into the melt glass that coalesces at the base of the
15 cavity. However, refractory radionuclides can also be distributed more broadly in the cavity
16 region as fine droplets become entrained with escaping cavity gases or as a result of splashing if
17 the melt glass is still molten when rubble in the cavity–chimney area collapses. Volatile species
18 with lower boiling points (e.g., tritium, Cl, I, alkalis, Ru, U, Sb) circulate up cracks in the rubble
19 chimney. As the cooling progresses, the melt begins to quench, and elements with lower boiling
20 points subsequently condense onto exposed mineral and fracture surfaces within the cavity and
21 the collapsed rubble. A portion of the volatile species can also be volumetrically incorporated
22 into any late-stage melt residual. Tritium will condense as molecular HTO together with the
23 large amounts of steam produced by the explosion. Fractions of volatile radionuclides with
24 higher solubilities (e.g., ^{36}Cl and ^{129}I) will also be partitioned into the condensed water. Gas

1 species (e.g., ^{90}Kr and ^{137}Xe) are transported through the rubble and will be concentrated higher
2 in the cavity and in the chimney relative to the refractory radionuclides.

3 In tests conducted beneath the water table, the condensed water will mix with
4 groundwater invading from the periphery and occupy the interstitial voids of the rubble and glass
5 matrices. This will allow the “rubble-”, “water-”, as well as some of the “gas-based” fractions of
6 the inventory to become incorporated in and mobilized by the groundwater, and for the “glass-
7 bound” fractions to slowly dissolve into the groundwater over the long term.

8 **3. Data analyses**

9 NTS “hot” (near-field) wells refer to monitoring wells where the samples contain
10 radionuclides above safe-drinking-water standards, defined on the basis of tritium, in the state of
11 Nevada. A number of hot wells were completed opportunistically in post-shot re-entry boreholes
12 drilled into a nuclear test cavity or chimney for diagnosing device performance. Due to cavity
13 collapses and well plugging, only 22 out of nearly 300 saturated nuclear events are available for
14 sampling. The body of data accumulated over 30 years from these wells is used to assess the
15 solubility and mobility of radionuclides at NTS.

16 A database was compiled to contain radionuclide concentration data from 18 monitoring
17 wells for 14 nuclear tests at NTS, with a total of 1285 sampling events (Table 2). We focus on
18 the measured activity for ^3H , ^{14}C , ^{36}Cl , ^{99}Tc , ^{129}I , ^{90}Sr , ^{137}Cs , ^{237}Np , and uranium and plutonium
19 isotopes (combined $^{239+240}\text{Pu}$ value for general public release), as available, that are relevant to
20 Yucca Mountain repository. These activities were decay-corrected to September 23, 1992, the
21 date of the last underground nuclear test at NTS. By this decay correction, we can directly
22 compare the measured activity with radionuclide residual inventory from all nuclear tests at NTS
23 (Smith et al., 2003). The inventory available to the general public sums the radionuclide totals

1 for individual tests into six principal geographic test areas; the test-specific inventory is
2 classified. This geographic averaging process may minimize the contribution from any single
3 test, particularly for higher-yield tests. As seen in Figure 2, which compares the radionuclide
4 concentration ratio to tritium at geographic centers and the whole NTS, the geographic averaging
5 provides a reasonably consistent ratio, especially the observed small variability for ^{99}Tc and ^{129}I
6 that are of particular interest in this study. However, it must be noted that the radiologic source
7 information, which is not open to the public, is device-dependent.

8 Because tritium is probably the best tracer for tracking water flow and radionuclide transport
9 (Davis et al., 1980), we used a ratio approach to assess the migration of other radionuclides, with
10 tritium as the reference nuclide (i.e., $^{99}\text{Tc}/^3\text{H}$ and $^{129}\text{I}/^3\text{H}$). Dependent upon the location
11 (chimney-cavity or satellite region) of a monitoring well, either a “distribution ratio” or
12 “migration ratio” is obtained (Figure 3). This radionuclide ratio was further normalized by
13 dividing it with the “radiologic source ratio” of the source-term inventory for the principal
14 geographic test center wherein the nuclear test is located. This normalization provides us with an
15 estimate of the relative radionuclide distribution or migration behavior, compared to tritium. In
16 other words, if a radionuclide behaves like tritium in its initial distribution (Table 1) or
17 subsequent migration, the normalized concentration ratio will be 1. As a portion of the near-field
18 wells sample the cavity–chimney region (Table 2), the normalized ratio value of radionuclides
19 (“distribution ratio” divided by the “radiologic source ratio”) will shed light on their distribution
20 in the aqueous, as opposed to the nonsoluble, phase; a ratio significantly lower than 1 indicates
21 that this radionuclide (e.g., Pu) exists predominantly in nonsoluble (glass and rubble) phases.
22 Some monitoring wells are satellite wells located a certain distance (up to several hundred
23 meters) away from the cavity–chimney region, and the normalized ratio (“migration ratio”

1 divided by the “radiologic source ratio”) helps us understand the migration behavior of
2 radionuclides; a ratio of 1 suggests the radionuclide migrates conservatively like tritium.

3 To evaluate the approach of using the area-averaged radionuclide ratio to assess the
4 radionuclide migration at NTS, Table 3 presents the ratio comparison between CAMBRIC test,
5 the only published source inventory for certain radionuclides, and the Frenchman test area
6 wherein CAMBRIC test occurred. In this case, the difference between using the actual and the
7 non-test-specific radionuclide source inventories is no more than a factor of 7. Also presented in
8 Table 3 are the ratios for these radionuclides in all the underground nuclear tests detonated at
9 NTS. Note that the Frenchman test area has the fewest nuclear detonations (10) among all 6
10 geographic test centers. For the majority of radionuclides, the difference between all geographic
11 test centers and the NTS total is within a factor of 3; comparison for some radionuclides is
12 presented in Figure 2. Overall, this study provides a qualitative assessment of the field-scale
13 behavior of some presumably conservative species.

14 It should be emphasized that each hot well is unique and represents a wide range in
15 geologic and hydrologic conditions, radiologic source terms, and groundwater sample quality.
16 Significant differences in sampling protocols have existed over the years, and caution has been
17 exercised when comparing data from different wells and factored in the discussion of this work.
18 Samples that were collected using downhole submersible pumps are more likely to be
19 representative of the bulk fluid composition in the near-field environment. Several of the wells,
20 particularly those that have been converted from post-shot re-entry holes, consist of a single 7 cm
21 carbon steel piezometer tube. In most cases, these wells have only been accessible using
22 wireline bailers. Given that the fluid in the piezometer tube cannot freely circulate, samples

1 collected from these sites tend to be of lower quality than those produced from pumped wells.
2 Sampling methods are noted in Table 2.

3 **3. Results and Discussion**

4
5 We now proceed to discuss the fate and transport behavior of important radionuclides,
6 analytical methods, and present the results obtained from the synthesis of available radionuclide
7 concentration data at NTS. This work focuses on the presumably mobile radionuclide species,
8 such as ^3H , ^{14}C , ^{36}Cl , ^{99}Tc and ^{129}I . The discussion centers on the comparison between
9 normalized ratios among samples collected from cavity–chimney and the satellite wells.

10 11 3.1. *Tritium*

12 Tritium (half-life [$t_{1/2}$] is 12.26 years) is the most abundant radionuclide produced during
13 underground testing, and can be measured by liquid scintillation counting. Following a nuclear
14 detonation, tritium occurs mainly in the forms of hydrogen gas (HT) and tritiated water (HTO).
15 Tritium is probably present in free-radical form at early times after the nuclear explosion. The
16 tritium free radicals are mixed intimately with the vaporized water at temperatures sufficiently
17 high to cause dissociation of the water molecules and to allow incorporation of the tritium in
18 some of them. Ninety-eight percent of the tritium becomes associated with water vapor shortly
19 after detonation and then condenses into the liquid phase, leaving only 2% in the form of HT
20 (Table 1).

21 Tritium has been considered an ideal water tracer (Davis et al., 1980). However, a small
22 yet noticeable retardation of tritium in transport studies has been reported in numerous
23 publications (e.g., Wierenga et al., 1975; van Genuchten and Wierenga, 1977; Gaber et al., 1995;
24 Hu and Brusseau, 1996). The retardation factor for tritium was measured to be 1.17 in an
25 aggregated tropical soil (Seyfried and Rao, 1987) and 1.2 in a column packed with glass beads

1 and porous kaolinite spheres (Hu and Brusseau, 1996). Seyfried and Rao (1987) also reported a
2 significantly greater tritium sorption from batch adsorption experiments than column transport
3 studies. A linear sorption isotherm of tritium was observed, with K_d values of 0.133 ± 0.0046
4 mL/g from batch experiments, compared to K_d values of 0.0653 ± 0.0044 mL/g from column
5 studies.

6 The interaction of tritium with solid surfaces has been postulated to occur via isotope
7 hydroxyl exchange with clay lattice hydroxyls (Stewart and Baker, 1973). Because of the low
8 mass numbers of hydrogen isotopes, the differences in their atomic masses and zero-point
9 vibrational energy bonds are large, and there is a high potential for an isotopic effect for tritium
10 (Stewart, 1972). This exchange is more pronounced in kaolinite minerals that have available
11 hydroxyl groups at platelet surfaces. In contrast, montmorillonite and illite minerals undergo
12 exchange only at the edges of the plates (where the hydroxyl groups are available), and thus
13 isotopic exchange is almost negligible (Gvirtzman and Magaritz, 1986). Stewart (1970)
14 suggested that the interaction between tritium and solid surfaces depends upon moisture content
15 and will be greater during the flow of tritiated water into initially dry soil than during miscible
16 displacement flow in saturated soils.

17 An appreciable sorption of tritium is not expected at NTS because the geological media at
18 NTS do not contain the high amount of clays needed for tritium retardation, except for rocks
19 with appreciable amount of zeolite minerals (Rose et al., 2000a). However, Burbey and
20 Wheatcraft (1986) reported that invocation of tritium sorption was necessary to match the tritium
21 elution curve at the CAMBRIC migration test. Bryant (1992) cautioned that (1) the model fit
22 with no tritium sorption was quite good and (2) the magnitude of the reported K_d value (0.8
23 mL/g) by Burbey and Wheatcraft (1986) is unusually large. Nevertheless, Bryant (1992)

1 speculated that although the retardation effect is relatively small, it may not be zero, which is
2 consistent with literature publications discussed above.

3 3.2. Carbon-14

4 ^{14}C ($t_{1/2}$ is 5,715 years) is expected to exist as the bicarbonate anion in NTS groundwater.
5 Before 1998, ^{14}C was measured by liquid scintillation counting; more recently, it has been
6 measured by accelerator mass spectrometry (AMS) with low detection limit and high accuracy.
7 Most normalized $^{14}\text{C}/^3\text{H}$ ratios are close to 1 at the cavity–chimney wells (Figure 4a), indicating
8 that ^{14}C exists mostly in the aqueous phase in the initial distribution after nuclear tests; this is
9 consistent with its expected behavior (Table 1). ALMENDRO samples consistently show lower
10 ratios than other cavity-chimney wells; this well is postulated to be hydraulically isolated as
11 discussed in Section 3.3. For the DALHART and BOURBON tests, ^{14}C data exhibit variability
12 (more than one order of magnitude) among samples collected at different durations (Figure 4a).
13 The first sample shown in Fig. 5a at the DALHART well was bailed in 1997, and the later three
14 samples were pumped but only a small ($< 27 \text{ m}^3$) amount of fluid could be pumped out before
15 sampling; all the samples may not be reproducibly collected at different times. Sample variability
16 might exist for the BOURBON results as well, when the two available ^{14}C samples were
17 collected by bailing in 2001 and 2005.

18 The normalized ratios for ^{14}C at most satellite wells are similar to those at cavity–
19 chimney wells. This suggests that dissolved ^{14}C migrates *qualitatively* similarly as tritium in
20 saturated groundwater of volcanic and carbonate rock. Nevertheless, the only pair of cavity–
21 satellite wells with available ^{14}C data is from the CAMBRIC test, and indicates 3 orders of
22 magnitude difference between the cavity (RNM-1) and satellite well (RNM-2S) well 91 meters
23 away. Furthermore, the ^{14}C ratio from the far field well (UE-5n) is also found to be 10 times

1 lower than its discharge source (RNM-2s) at the CAMBRIC vadose zone test (Tompson et al.,
2 2002). The retarded transport of ^{14}C is most likely related to its isotopic exchange with non-
3 mobile carbon atoms in the aquifer material, such as those of carbonate. Retardation of ^{14}C is
4 much more noticeable in the alluvium aquifer than in volcanic aquifers.

5 3.3. Chlorine-36

6 ^{36}Cl ($t_{1/2}$ is 3.01×10^5 years) is expected to exist as the chloride anion in NTS groundwater.
7 For NTS groundwater samples, the atom ratio of $^{36}\text{Cl}/\text{Cl}$ is measured by AMS, and the
8 concentration of ^{36}Cl is calculated using the stable chloride concentration from ion
9 chromatography. The surface charge for many clay minerals of the geologic media is
10 predominantly negative and repels the negative charge carried by anions. As a result, chloride is
11 not expected to sorb onto geologic media. On the contrary, anion exclusion could occur when
12 anions are partially or totally excluded from some flow paths, resulting in a greater average
13 velocity for anions (such as chloride) than for neutral water molecules. For a soil that contained
14 40% clay, Thomas and Swoboda (1970) found that the rate of chloride movement was 1.37 times
15 faster than that of deuterium. During the CAMBRIC pumping experiment, the breakthrough and
16 the maximum concentration of ^{36}Cl appeared slightly ahead of tritium in alluvium (Ogard et al.,
17 1988). They attributed this to the anion exclusion effect that effectively prevented $^{36}\text{Cl}^-$ from
18 entering the intragranular porosity of the soil particle. The calculated anion exclusion volume of
19 the alluvium at NTS was 7.0 mL of liquid per 100 g of soil (Ogard et al., 1988).

20 Here we point out that both early (anion exclusion) and delayed (anion sorption) transport
21 of anions have been reported in various geologic media (e.g., McMahon and Thomas, 1974;
22 Chan et al., 1980; Gvirtzman et al., 1986; Toner et al., 1989; Ishiguro et al., 1992; Seaman, 1998;
23 Katou et al., 2001; McVay et al., 2004; Hu et al., 2005). Bresler (1973) proposed an empirical

1 relationship to evaluate the relative exclusion concentration, as a function of $b\sqrt{C}$ (where b is
2 the water-film thickness in Å and C is the total anion concentration in eq/L). The process and
3 magnitude of anion exclusion/sorption are related to the nature of the clay minerals, clay content,
4 liquid saturation, iron oxide content, and anion concentrations (Thomas and Swoboda, 1970;
5 Appelt et al., 1975; James and Rubin, 1986). Without complete information for these parameters,
6 it is difficult to ascertain whether anion exclusion or anion sorption is operative. For example,
7 McMahon and Thomas (1974) observed an anion exclusion effect for chloride transport,
8 compared to tritium, in two of three soils investigated and observed chloride sorption in another
9 soil. The difference is ascribed to the amount of positively charged iron oxides and to anion
10 exchange capacity.

11 Apart from the CAMBRIC forced-gradient migration test discussed in Ogard et al. (1988)
12 for ^{36}Cl , Figure 4b presents the available ^{36}Cl at the NTS. The distribution ratios (i.e., $^{36}\text{Cl}/^3\text{H}$
13 ratio for the cavity-chimney wells) are all less than 1, indicating different behavior for ^{36}Cl and
14 ^3H . This is consistent with the partitioning estimated in an IAEA study (1998) which suggested
15 values of 50% in melt glass, 40% in rubble, and 10% in groundwater. Qualitatively, the satellite
16 wells have $^{36}\text{Cl}/^3\text{H}$ ratios that are similar to the cavity–chimney wells. This is especially shown in
17 BIBLY and CAMBRIC results where samples from both types of wells are available. The 2000
18 sample (measured ^{36}Cl activity at 1.59×10^{-5} Bq/L at the collection time) at RNM-1 well of
19 CAMBRIC test has a much lower $^{36}\text{Cl}/^3\text{H}$ ratio than the 2004 sample (^{36}Cl activity at 1.64×10^{-5}
20 Bq/L), and the different $^{36}\text{Cl}/^3\text{H}$ ratio lies in the different ^3H activity (1,051 in 2000 vs. 12.6 Bq/L
21 in 2004). In other words, ^3H is moving away from the CAMBRIC cavity faster than ^{36}Cl under
22 the natural gradient during 2000-2004, if the 2000 sample was not an outlier. The 2004 cavity
23 sample is quite comparable with the satellite well samples at CAMBRIC test; more sampling at
24 the RNM-1 will resolve this issue.

1 3.4. Technetium-99

2 ^{99}Tc is a long-lived ($t_{1/2} = 2.13 \times 10^5$ years), abundant (6% yield) fission product of ^{235}U
3 and ^{239}Pu . NTS hot-well water samples have been measured for ^{99}Tc by radiochemical counting
4 (Silva et al., 1988). Some NTS samples were analyzed for ^{99}Tc by AMS, which provides much
5 greater sensitivity and precision than conventional radiochemical counting (Bergquist, et al.,
6 2000). Recently, we have developed a routine and robust method to concentrate, purify and use a
7 quadrupole ICP-MS to analyze ^{99}Tc in NTS groundwaters.

8 Depending upon the redox conditions, Tc primarily exists in two stable oxidation states.
9 It forms a reduced species [predominantly Tc(IV)] at redox potential (Eh) values below about
10 220 mV with respect to standard hydrogen electrode (SHE) in neutral pH conditions. At higher
11 Eh, it occurs as Tc(VII)O_4^- . Due to its weak interaction with mineral surfaces, TcO_4^- is deemed
12 one of the most mobile radionuclides in the environment. In contrast, transport of Tc(IV) species
13 ($\text{TcO}_2 \cdot n\text{H}_2\text{O}$) are expected to be strongly retarded from their interactions with minerals and/or
14 precipitation due to their low solubility; the solubility of $\text{TcO}_2 \cdot n\text{H}_2\text{O(s)}$ in carbonate-containing
15 groundwater was reported to be about 10^{-8} M (Eriksen et al., 1992) . From experiments
16 conducted at five sediments and groundwaters in Germany, a dramatically different sorption
17 coefficient for Tc under both aerobic and anaerobic conditions was obtained (Lieser and
18 Bauscher, 1987). By varying the redox potential, they observed a drastic drop in the K_d value for
19 Tc by about 3 orders of magnitude within a small range of Eh at 170 ± 60 mV for a pH of $7 \pm$
20 0.5. Chemical equilibrium modeling using EQ3/6 software indicated Tc solubility of 4×10^{-9} M
21 under reducing conditions in the saturated zone at Yucca Mountain, and transition from lower to
22 higher solubility ranges from -100 mV to about +300 mV/SHE (Arnold et al., 2006).

23 From the CAMBRIC pumping test, Schroeder et al. (1993) reported that ^{99}Tc in solution
24 appears to migrate a bit more slow than ^{36}Cl . The initial breakthrough for ^{99}Tc is similar to

1 tritium; the migration rate, evaluated from the center of mass, of ^{99}Tc appears to slightly exceed
 2 that of ^3H , perhaps as a result of anion exclusion effects. However, this discussion was based on
 3 a very limited number (one in RNM-1S, and 3 in RNM-2S) of ^{99}Tc data. They also reported a
 4 $^{99}\text{Tc}/^3\text{H}$ ratio of 2.8×10^{-7} for the CAMBRIC source term and an activity ratio of 3.4×10^{-11}
 5 measured in the cavity water. This indicates that only 0.01% of the ^{99}Tc source term was in the
 6 aqueous phase and available for transport; the remainder is presumably contained in the melt
 7 glass.

8 Apart from the ALMENDRO test which has persistently exhibited a reducing cavity-
 9 chimney environment (to be discussed in more detail in Section 3.5) and associated reduced
 10 redox state and solubility of Tc, available $^{99}\text{Tc}/^3\text{H}$ ratios in all NTS cavity-chimney wells are
 11 mostly in the range of 0.1 to 1. This indicates that up to 10% of ^{99}Tc (compared to 98% for ^3H)
 12 produced from nuclear tests is present in the aqueous phase. This value is not too far from the
 13 estimated partitioning of ^{99}Tc by the IAEA (1998), which suggested that 20% exists in the
 14 relatively accessible rubble phase (Table 1). The possibility that a large proportion (80%) of Tc
 15 could be incorporated into the melt glass is due to the fact that the parent chain of ^{99}Tc includes
 16 ^{99}Zr ($t_{1/2} = 2.2$ s), ^{99}Nb ($t_{1/2} = 15.0$ s), and ^{99}Mo ($t_{1/2} = 2.75$ days; which likely matters the most
 17 because of its relatively long half-life that allows a significant fraction of mass 99 to partition
 18 into the glass), all of which are relatively refractory.

19 During the pumping test, the $^{99}\text{Tc}/^3\text{H}$ ratio was similar for wells RNM-1 and RNM-2s,
 20 suggesting that the soluble ^{99}Tc migrated as rapidly as the tritiated water in the oxygenated water
 21 at CAMBRIC site (Bryant, 1992). ^{99}Tc may also occur predominantly as TcO_4^- at CHESHIRE
 22 test, as it showed very constant concentration ratios to tritium in the cavity, formation, and
 23 satellite measurements (Buddemeier et al., 1991). Apart from the CAMBRIC and CHESHIRE
 24 data, the $^{99}\text{Tc}/^3\text{H}$ ratios at three other available satellite wells are nearly 2 orders of magnitude

1 lower than the cavity–chimney values (Figure 5). This indicates that field-scale migration of ^{99}Tc
2 at NTS is likely retarded and slower than that of tritium, with an apparent retardation factor of
3 about 100. This means that the TSPA analyses are probably too conservative by using a K_d value
4 of zero in tuff for ^{99}Tc , and natural barriers of NTS subsurface could yield performance of
5 retarding its transport to downgradient receptors. Delayed transport of ^{99}Tc is not unexpected in
6 the locally reducing groundwater at NTS.

7

8 *3.5. Redox chemistry of cavity and NTS groundwater*

9 To understand the non-conservative transport of ^{99}Tc , we need to consider the redox
10 chemistry of both cavity water and local groundwater at NTS. Several lines of evidence from
11 NTS strongly suggest that the cavity environment is likely reducing, or that the cavity
12 environment was reducing at least during the early time after nuclear explosions and may last for
13 some duration of time. The study of Borg (1975) directly pointed to a reducing cavity
14 environment, at least at the time when melt glasses are formed. Chemical analyses of the melt
15 glasses from RAINIER test in 1957 and PILED RIVER test in 1966 showed the conversion of
16 ferric iron (Fe^{3+}) to ferrous (Fe^{2+}) ion, which indicated that a reducing atmosphere prevailed in
17 the cavities at the time of quenching.

18 Whether the test cavity remains reducing depends upon its hydraulic connection to the
19 groundwater. While test cavities such as CAMBRIC and CHESHIRE have been oxidizing, our
20 studies of ALMENDRO, CAMEMBERT and CHANCELLOR tests indicate a reducing cavity
21 environment persists due to poor circulation. Anomalously enriched $\delta^{13}\text{C}$ values in
22 ALMENDRO groundwater samples suggest that methanogenesis process is occurring (Rose et
23 al., 2000b). The reduction of CO_2 to CH_4 is associated with progressively enriched ^{13}C over time
24 in a closed system. Analysis of a gas sample, collected 107 m below the surface in the

1 CAMEMBERT post-shot hole, showed that the gas, relative to normal air, is significantly
2 depleted in oxygen ($0.12 \pm 0.01\%$) and enriched in hydrogen gas ($8.28 \pm 0.01\%$). This is
3 characteristic of the anoxic, reducing condition favorable to methanogenesis; the gas was found
4 to contain 1% methane and no CO_2 . For methanogenesis to occur, the redox potential is expected
5 to be lower than -244 mV/SHE (at pH 7), which would indicate a strongly reducing environment
6 at these tests.

7 Similar to the initial reducing condition of radiologic source in an underground nuclear
8 test, the spent nuclear fuel to be stored at Yucca Mountain is in a reduced condition, with
9 reduced uranium in the form of UO_2 comprising of more than 95% of the total radioactivity
10 (Ewing and Macfarlane, 2002). If the engineered barriers fail, UO_2 would not be stable in the
11 presence of even minor amounts of moisture and under oxidizing environments at the vadose
12 zone of the repository horizon. Under Eh-pH conditions expected for the oxidative corrosion of
13 spent nuclear fuel, TcO_4^- will be the predominant species of technetium with $\log [\text{TcO}_4^- / \text{TcO}_2 \cdot$
14 $\text{nH}_2\text{O}] > 2.15$ in the pH range of 4 to 10 (Chen et al., 2000).

15 Below the water table, where gas exchange with the atmosphere diminishes, dissolved
16 oxygen (DO) is gradually consumed by microbial uptake, biodegradation of organic matter, and
17 retention on reduced mineral phases in the aquifer. It has often been assumed that deep aquifers
18 ($>250 \text{ m}$) are reducing since they are isolated from the atmosphere. Although the water table at
19 NTS ranges from 400 to 700 m deep, the groundwater has been postulated oxygenated
20 (Winograd and Robertson, 1982; Coles and Ramspott, 1982; Buddemeier and Hunt, 1988).
21 Nevertheless, this is based on limited information. Winograd and Robertson (1982) reported that
22 even very old groundwaters ($>10,000$ years), along an approximately 80-km flow path from
23 recharge areas in the Paleozoic carbonate-rock aquifer of the Ash Meadows groundwater basin
24 (south-central Nevada) near NTS, contain DO concentrations close to air saturation.

1 Since 2000, Finnegan and his coworkers (Finnegan and Thompson, 2005) employed a
2 Hydrolab Minisonde 4a multi-probe to measure several geochemical parameters (oxidation-
3 reduction potential, DO, temperature, pH, and specific conductivity) in situ in non-radionuclide-
4 contaminated wells at NTS. From a total of 22 wells, a wide range of oxidation-reduction
5 potential (ERP; redox potential) and DO values were observed. The measured redox potential
6 indicates that water is oxidizing (redox potential ranges from about 200 to 400 mV) in seven
7 wells, mildly reducing (about -100 to 200 mV) in seven wells, and reducing (from -300 to -100
8 mV) in eight wells. Note that the measured redox potentials were not corrected to SHE values
9 (Eh; probably add 200 to the redox potential to convert to Eh), yet either potential can serve as a
10 qualitative indicator of redox conditions of the groundwater. Overall, the ERP values correspond
11 with the DO values, and decrease with the increasing depth of measurement below the water
12 table (examples shown in Figure 6). The occurrence of observed reducing groundwater seems to
13 be randomly distributed (no correlation with well locations) at NTS, and the reducing
14 groundwater is observed in the northern and north-western area of NTS (Figure 7). However,
15 there are some concerns about whether these measurements in non-purged wells represent the
16 ambient conditions of NTS groundwater. The downhole probe could only measure stagnant
17 water and, furthermore, could be compromised by the potential corrosion of steel casing in some
18 well bores. Information about well construction, completion, and geochemical monitoring data is
19 limited to evaluate the representativeness of localized reducing groundwater.

20 Meijer and coworkers have been examining variations in the redox potential in
21 groundwaters pumped from 20 boreholes in the saturated zone along potential transport
22 pathways from Yucca Mountain (SBC, 2004). At least three borehole volumes of groundwater
23 were purged from the borehole prior to the sampling the water. Redox conditions in groundwater
24 are characterized by the measurement of various parameters including oxidation/reduction

1 potential (ORP/Eh), dissolved oxygen, and the concentrations of redox-sensitive constituents
2 such as ferrous iron, total iron, total manganese, nitrate, nitrite, ammonium, sulfide, sulfate, as
3 well as other less commonly analyzed constituents such as different oxidation states of antimony,
4 arsenic, and selenium. The boreholes in which reducing conditions have been found in
5 groundwater include those on or near Yucca Mountain, the borehole in alluvium to the east of
6 Fortymile Wash, and a group of boreholes to the west of Fortymile Wash near Yucca Mountain.
7 The boreholes on Yucca Mountain that contain reducing waters are relatively deep boreholes that
8 penetrate the Tram Member of the Crater Flat Tuff. This member includes a volcanic unit that
9 contains pyrite (FeS_2), which is a likely source of the reducing conditions. It is important to
10 recognize that the presence of reducing conditions in more than one borehole located directly to
11 the east of the repository footprint suggests there may be a volume of rock east of the repository
12 footprint that contains reducing groundwater. This volume (reducing curtain) could be a
13 substantial barrier to the transport of redox-sensitive radionuclides in the saturated zone. The
14 groundwaters showing reducing conditions in boreholes to the east and west of Fortymile Wash
15 near the southern boundary of the site include boreholes known to contain pyrite based on core
16 descriptions. The other boreholes may also contain pyrite although core descriptions are not
17 available to test this possibility (SBC, 2004).

18 In addition to the reducing groundwater, the presence of reductants (e.g., Fe(II) and S^{2-})
19 can also contribute to the reduction of Tc(VII) to Tc(IV) . Fe(II) minerals in igneous rocks can
20 reduce TcO_4^- and lead to sorption on mineral surfaces (Bondietti and Francis, 1979). Under
21 reducing conditions, Cui and Eriksen (1996b, 1998) discovered that TcO_4^- was reduced to
22 sparingly soluble $\text{TcO}_2 \cdot n\text{H}_2\text{O(s)}$ by the Fe(II) -containing fracture-filling material and that
23 $\text{Tc(IV)}_{\text{aq}}$ was rapidly sorbed by the material. Reduction of Tc(VII) to Tc(IV) occurs with Fe(II) -
24 containing solid phases but not by aqueous Fe(II) species (Cui and Eriksen, 1996a). During the

1 CHESHIRE migration test, ^{99}Tc concentration was observed to drop precipitously for
2 groundwater samples with high iron content, while ^{36}Cl was not affected (Buddemeier et al.,
3 1991). No explanation was offered, and it seems probable that the reduction of Tc(VII) to Tc(IV)
4 and the formation of an insoluble precipitate constitute the likely mechanisms for the much
5 slower mobility of ^{99}Tc .

6 Based on above discussion of localized reducing groundwater and mineral-mediated
7 reduction, it is likely that Tc will not always exist as TcO_4^- and act as a mobile radionuclide in
8 the groundwater of NTS. Numerical simulations show the enhanced sorption within a reducing
9 zone of modest width leads to significantly greater retardation of redox-sensitive radionuclides
10 (^{99}Tc and ^{237}Np) in the saturated zone (Arnold et al., 2006).

11 3.5. Iodine-129

12 Similar to ^{99}Tc , ^{129}I has a long half-life ($t_{1/2} = 1.57 \times 10^7$ years) as well as a unique and
13 complex chemistry in the environment. (The fission product ^{129}I in NTS well data has been
14 measured by neutron activation, thermal-ionization mass spectrometry, and AMS; all samples
15 discussed in this work were analyzed by AMS.) The fate and transport of ^{129}I in groundwater are
16 dictated by its chemical speciation. Aqueous iodine usually occurs as the highly mobile iodide
17 anion (I^-). Under more oxidizing conditions, iodine may be present as the more reactive iodate
18 anion (IO_3^-); its interaction with clays and organic matter leads to its retarded transport (Couture
19 and Seitz, 1983; Sheppard and Thibault, 1992). Different from other redox-sensitive
20 radionuclides (such as ^{99}Tc), iodine has a strong retardation under oxidizing conditions.
21 Furthermore, coexistence of several iodine species has been reported in various aqueous systems
22 (cf., Hu et al., 2005).

1 Although a substantial fraction of the ^{129}I observed in the RNM-1 cavity well migrated to
2 the RNM-2s satellite well at the CAMBRIC test, some of the ^{129}I is immobile or its migration is
3 retarded to some extent (Bryant, 1992). It was postulated that reactive HIO is the predominant
4 species and that it may interact with matrix materials. This work also confirms that ^{129}I is
5 migrating slower than ^3H , with a lower $^{129}\text{I}/^3\text{H}$ ratio in RNM-2S than in RNM-1; though the
6 results from the same well are somewhat variable (Figure 8). Measurements from other hot
7 wells exhibit no difference between cavity and satellite well samples. CHESHIRE is the only test
8 for which both cavity and satellite well samples are available, and the results show nearly the
9 same ^{129}I values. The normalized $^{129}\text{I}/^3\text{H}$ ratio value is abnormally large (nearly 10) for the
10 BOURBON and NASH tests; these samples were either sampled by bailing or after pumping
11 with a limited volume. Overall, available data show that it is likely that iodine species is
12 migrating conservatively at NTS as tritium.

13 However, it would not be surprising if iodine migration were retarded, since portion of
14 the NTS groundwater is strongly oxidizing, and the presence of high-valence-state iodine species
15 leads to their interactions with geological media. In a leachate of a tuff sample collected from an
16 underground nuclear explosion in hole U7ajs, Wolfsberg (1978) reported the existence of at least
17 three iodine species: IO_4^- , IO_3^- , and I^- , with proportions of 10, 40, and 50%, respectively. This
18 leachate was further used for batch sorption–desorption studies using alluvium samples from the
19 CAMBRIC site, and a very high sorption value of 640 ± 300 mL/g was obtained.

20 We used a Dionex Corp. (Sunnyvale, CA) ion chromatography DX-600 system to
21 measure low (sub-ppb) concentrations of iodide (Hu et al., 2005). The pulsed amperometric
22 detector has a detection limit of 0.6 mg/L for iodide. Total iodine was analyzed by ICP-MS with
23 a detection limit about 0.1 mg/L. Total iodine is expected to be composed of both inorganic
24 iodine (i.e., iodide, iodate) and organic iodine. Currently there are no low-detection methods to

1 independently measure all of the potential inorganic iodine species and organic iodine. The
2 approach used here provides information about the proportion of total iodine existing as iodide
3 which is an indication of redox condition. For the oxygenated CAMBRIC sites, all 9 samples in
4 all 3 wells show less than 10% of iodine species existing as low-valence iodide. On the contrary,
5 iodide comprises more than 60% of total iodine in ALMENDRO, CAMEMBERT, and
6 CHANCELLOR wells which have been independently confirmed to be reducing. Overall results
7 show that 79% of samples has less than 50% of iodine in the form of iodide, among a total of 43
8 NTS groundwater samples analyzed for iodide and total iodine. This indicates that a considerable
9 amount of iodine may exist in non-conservative species. The implications of changing redox
10 state are comparatively less significant for I than for Tc, for which solubility and migration
11 potential are dramatically different between +7 and +4 valence states.

12

13 **4. Conclusions**

14 Because of the comparable hydrogeologic settings, understanding gained from NTS is
15 applicable to the performance evaluation of the proposed repository at Yucca Mountain. In this
16 study we synthesized a body of radionuclide concentration data accumulated over the past
17 several decades at NTS. The results provide an independent validation, though qualitatively, at
18 field scales over distances of hundreds of meters, of the sorption values used in TSPA analyses
19 for Yucca Mountain repository. Migration of ^{14}C at the NTS is likely conservative as tritium in
20 volcanic rock, yet some retarded transport from isotopic exchange might occur in alluvium
21 aquifer. ^{36}Cl transport at NTS is enhanced, compared to tritium, which is related to several
22 factors including anion exclusion, together with a slight delay of tritium transport from its
23 isotopic exchange with hydroxyl group in clay minerals.

1 Knowledge of the speciation and reactions of presumably mobile radionuclides, such as
2 long-lived ^{99}Tc and ^{129}I , is very important for understanding their transport behavior at Yucca
3 Mountain and NTS. Tc does not necessarily exist as the mobile and conservative species TcO_4^- ,
4 and the assumption of uniformly oxidizing groundwaters at NTS is partly based on the scarcity
5 of in situ water chemistry data in the subsurface. The retarded transport of ^{99}Tc is not unexpected
6 as the speciation and migration behavior of ^{99}Tc will be significantly controlled by the redox
7 conditions of the nuclear test-induced cavity (initial distribution) and surrounding groundwater
8 (subsequent migration). Emerging lines of evidence suggest that the chemical environment of a
9 nuclear test cavity is likely to have a reducing nature immediately following a detonation, similar
10 to the nuclear fuel before the breaching of the waste package. These conditions may persist for
11 decades after a test. Recent geochemistry measurements at wells across NTS and close to Yucca
12 Mountain indicate that groundwaters are not uniformly oxidizing, as previously believed. The
13 mobility of Tc (and Np) would be greatly reduced in a nonoxidizing environment. Similar to Tc,
14 the speciation and migration behavior of iodine are complex and dependent upon the redox
15 conditions, and different species have different reactivities and mobilities. Laboratory
16 experiments, performed under varying controlled redox states, are recommended to evaluate the
17 transport of redox-sensitive radionuclides (especially Tc, Np, and I) with representative
18 geological media and groundwater. Removal of the over-conservatism from presumably
19 conservative migration of these radionuclides (especially ^{99}Tc) will provide a realistic
20 performance of a geological repository.

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Table 1

Estimated percentage distribution of radionuclides among the glass, rubble, gas, and groundwater following a typical underground nuclear test (IAEA, 1998); data in parentheses for ^{90}Sr and ^{137}Cs are from Tompson et al. (2002).

Radionuclide	Glass	Rubble	Gas	Water
^3H			2	98
^{14}C		10	80	10
^{36}Cl	50	40		10
^{90}Sr	40 (25)	60 (75)		
^{99}Tc	80	20		
^{129}I	50	40		10
^{137}Cs	25(10)	75(90)		
U isotopes	90	10		
^{237}Np	95	5		
Pu isotopes	98	2		

Table 2 Compilation of nuclear tests, hot wells, and sample collections.

Test name	Test date	Working point below water table (m)	Yield † (kilotons)	Rock type [@]	Sampling well	Sample collection method	Sampling location with respect to the test hole	Sample collection dates (number) [#]
ALEMAN	9/11/1986	28	<20	VTA	UE-3e #4	Pump/Pressure tube	58 m north	1993 - 1998 (2)
ALMENDRO	6/6/1973	360	200-1000	TCU	U-19v PS#1 ds	Bailer	Cavity-chimney region	1993 - 2006 (8)
BILBY	9/13/1963	206	249	WTA	U-3cn PS#2	Pump	Chimney region	1977 - 2004 (15)
				LCA	U-3cn #5		129 m southeast	1980 - 1997 (3)
BOURBON	1/20/1967	-41	20-200	LCA	UE-7ns	Pump/Bailer	137 m away	1983 - 2001 (10)
BULLION	6/13/1990	53	20-150	TCU	ER-20-6 #1	Pump	166 m southwest	1996 - 1998 (6)
					ER-20-6 #2		207 m southwest	1996 - 1997 (4)
					ER-20-6 #3		296 m southwest	1996 - 1998 (4)
CAMBRIC	5/14/1965	77	0.75	AA	RNM-1	Pump	Cavity	1974 -2004 (25)
					RNM-2S		91 m from cavity	1975 - 1992 (950)*; 1978 - 2003 (14)
					UE-5n		510 m from RNM-2S	1999 - 2004 (3)
CAMEMBERT	6/26/1975	646	200-1000	TCU	U-19q PS#1 d	Pump	Chimney region	1998 - 2003 (2)
CHANCELLOR	9/1/1983	-20	143	LFA	U-19ad PS#1A	Pump	Cavity	2004 (1)
CHESHIRE	2/14/1976	536	200-500	LFA	U-20n PS#1 DDh	Pump	Cavity	1976 - 2005 (24)
					UE 20n-1		~ 300 m from cavity	1987 (98) [@] ; 1988 (3)
DALHART	10/13/1988	163	<150	TCU	U-4u PS#2a	Bailer (before 1997); Pump (after 1997)	Chimney region	1992 - 2003 (6)
GASCON	11/14/1986	103	20-150	TCU	UE-4t P#1	Pressure tube	~170 m to the south	1993 (1)
					U-4t PS#3a		~54 m from cavity wall	1992 (2)
INGOT	3/9/1989	-63	20-150	VTA	U-2gg PSE3a	Bailer	65 m from working point	1993 - 1994 (2)
NASH	1/19/1967	-163	39	LCA	UE-2ce	Bailer	183 m from working point	1982-1984 (65) ^{&} ; 1978 -2005 (26)
TYBO/BENHAM	5/14/1975	135	200-1000	WTA	ER-20-5 #1	Pump	280 m southwest	1996 - 2004 (5)
				LFA	ER-20-5 #3		280 m southwest	1996 - 2004 (6)

† Announced yields are from DOE/NV (2000).

@ VTA: vitric tuff aquifer; TCU: tuff confining unit; LCA: lower carbonate aquifer; AA: alluvium aquifer; LFA: lava flow aquifer; WTA: welded tuff aquifer.

The ER-20-5 well cluster was drilled in the near-field (~300 m from the surface ground zero) environment of the TYBO test. However, Pu isotopic signatures indicate the radionuclides in water samples from ER-20-5 wells are derived from the BENHAM test detonated ~1300 m up-gradient (Kersting et al., 1999).

The Radionuclide Migration Test was conducted at CAMBRIC, with continuous pumping during 1975-1992. These nearly 1,000 samples were analyzed for ^3H that is comprised of the majority of data, as well as ^{36}Cl , ^{85}Kr , ^{99}Tc , and ^{129}I , with the results discussed in Hoffman et al. (1977), Burbey and Wheatcraft (1986), Ogard et al. (1988), Bryant (1992), Schroeder et al. (1993), Tompson et al. (2002), and Guell and Hunt (2003).

@ Time-series samples at several sampling depths were collected from the pumping test at CHESHIRE site for the analyses of ^3H (mostly), as well as ^{36}Na , ^{85}Kr , ^{125}Sb , and ^{137}Cs ; results were published and discussed in Buddemeier and Hunt (1988), Buddemeier et al. (1991), and Sawyer et al. (1999).

& Periodic samples were collected during 1982-1984 at the NASH pumping test for the analyses of ^3H , ^{90}Sr , and ^{137}Cs (Daniels and Thompson, 1984).

Table 3 Comparison of ratios of various radionuclide activities to tritium, decay-corrected to September 23, 1992, among CAMBRIC test, Frenchman Flat, and NTS total.

Area \ Radionuclide	⁸⁵ Kr	⁹⁰ Sr	⁹⁹ Tc	¹²⁹ I	¹³⁷ Cs	²³⁸ U	²³⁹ Pu	²⁴¹ Am
CAMBRIC test †	1.10E-04	1.74E-03	1.32E-06	5.13E-09	5.18E-03	2.28E-06	1.50E-02	3.19E-03
Frenchman Flat	7.37E-04	1.08E-02	6.69E-06	2.60E-08	2.89E-02	5.45E-07	8.11E-03	2.88E-03
NTS total	1.42E-03	1.73E-02	4.54E-06	1.40E-08	2.27E-02	3.54E-07	1.27E-03	2.95E-04
Difference between Frenchman Flat area and CAMBRIC test	6.7	6.2	5.1	5.1	5.6	0.24	0.54	0.90

† Radiologic source term at the detonation date is from Hoffman et al. (1977) and Schroeder et al. (1993).

Figure Captions

- Figure 1 Map of the Nevada Test Site, and the potential Yucca Mountain repository, showing the near-field sampling sites. Both test name and associated near-field well (in parentheses) are provided. Numbers in subdivision are the area designations of NTS.
- Figure 2 Comparison of radionuclide ratio to tritium at six principal geographic test centers and the whole NTS.
- Figure 3 Illustration of hypothetical cross-section for nuclear test chimney-cavity and satellite sampling wells.
- Figure 4 Normalized concentration ratio for (a) ^{14}C and (b) ^{36}Cl in the hot wells. For a well with multiple sampling events, the data are shown with the latest sampling at the right-hand side.
- Figure 5 Normalized concentration ratio for ^{99}Tc in the hot wells. The radionuclide ^{99}Tc data with lower than detection limit are shown in a dashed circle.
- Figure 6 Measured in situ redox potentials at three representative wells at the Nevada Test Site: (a) oxidizing, (b) mildly reducing, and (c) strongly reducing. (Data from Finnegan and Thompson, 2001; 2002.)
- Figure 7 Redox conditions at 22 wells across the Nevada Test Site. Marker designation: circle – ERP ranging from about 200 to 400 mV; diamond – ERP about –100 to 200 mV; square – ERP from –300 to –100 mV. Enclosed contours indicate the principal geographic test centers, with the center name and number of underground nuclear test detonated in the center shown in the parentheses.
- Figure 8 Normalized concentration ratio for ^{129}I in the hot wells.

Figure 1

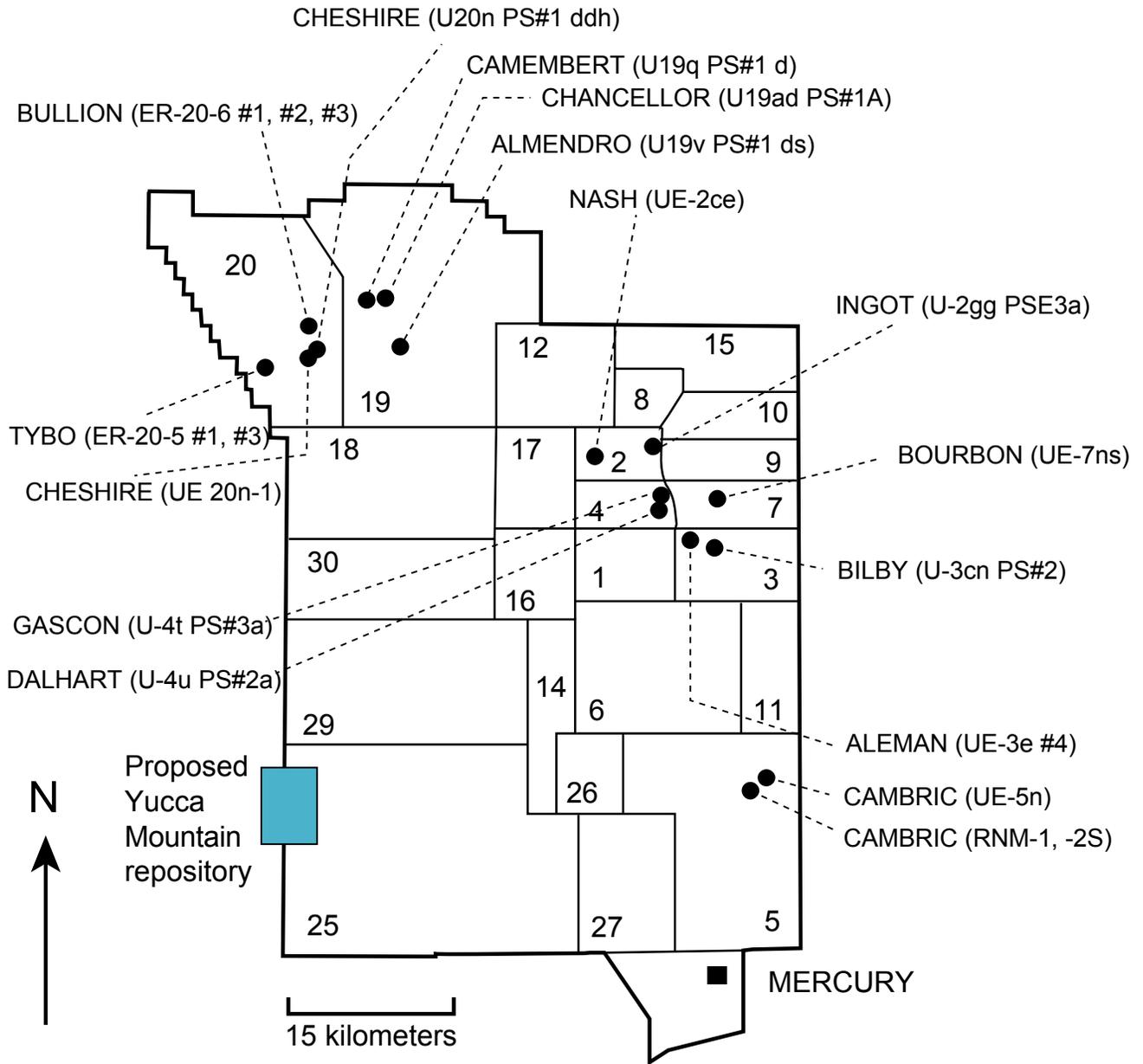


Figure 2

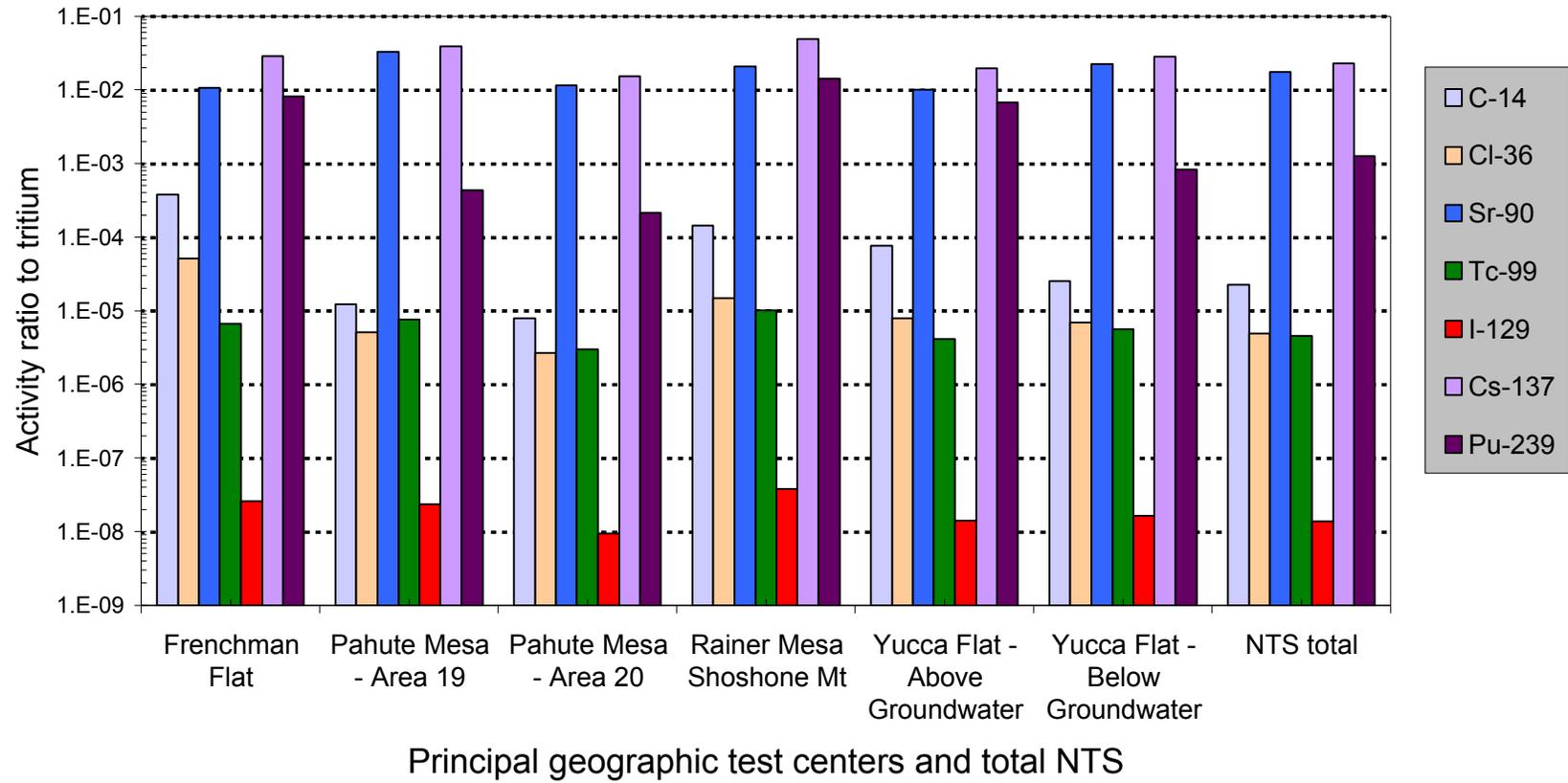


Figure 3

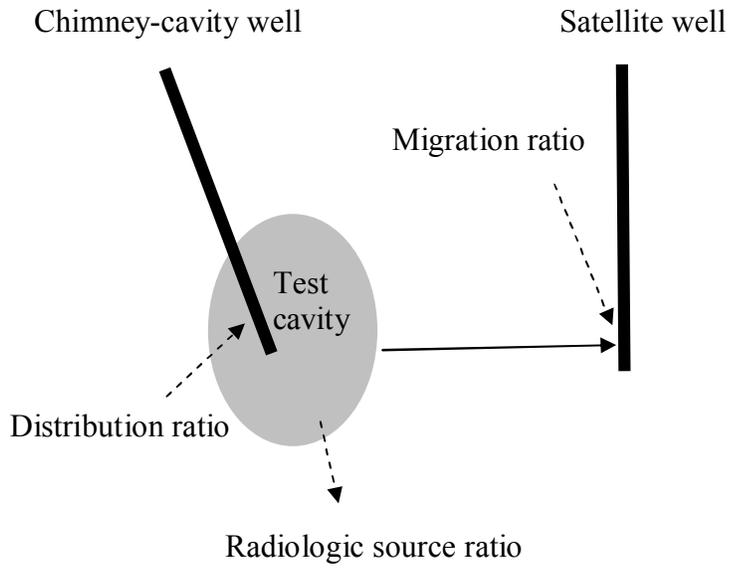
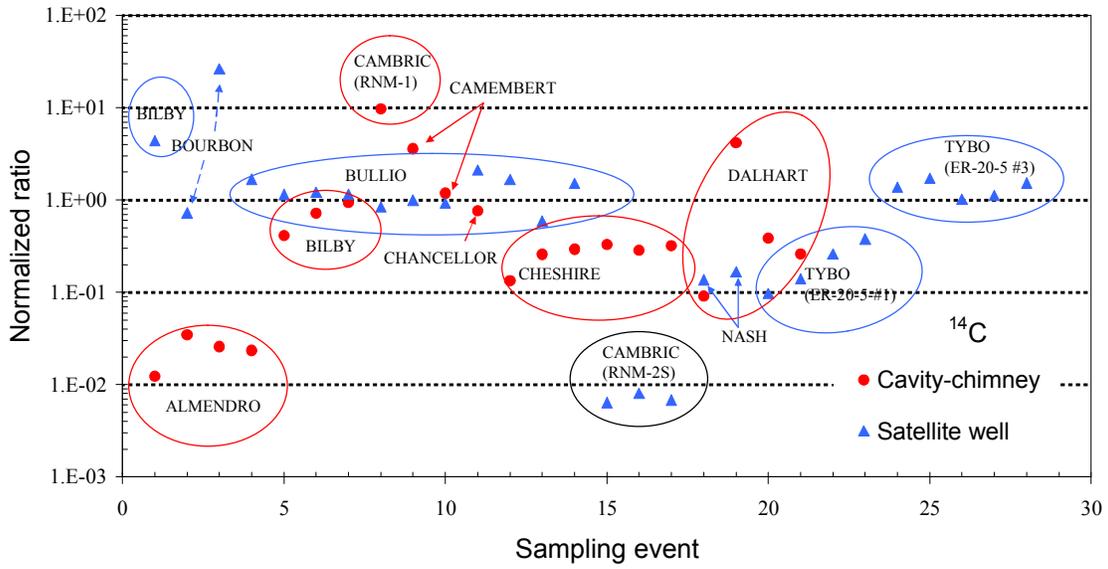


Figure 4

(a)



(b)

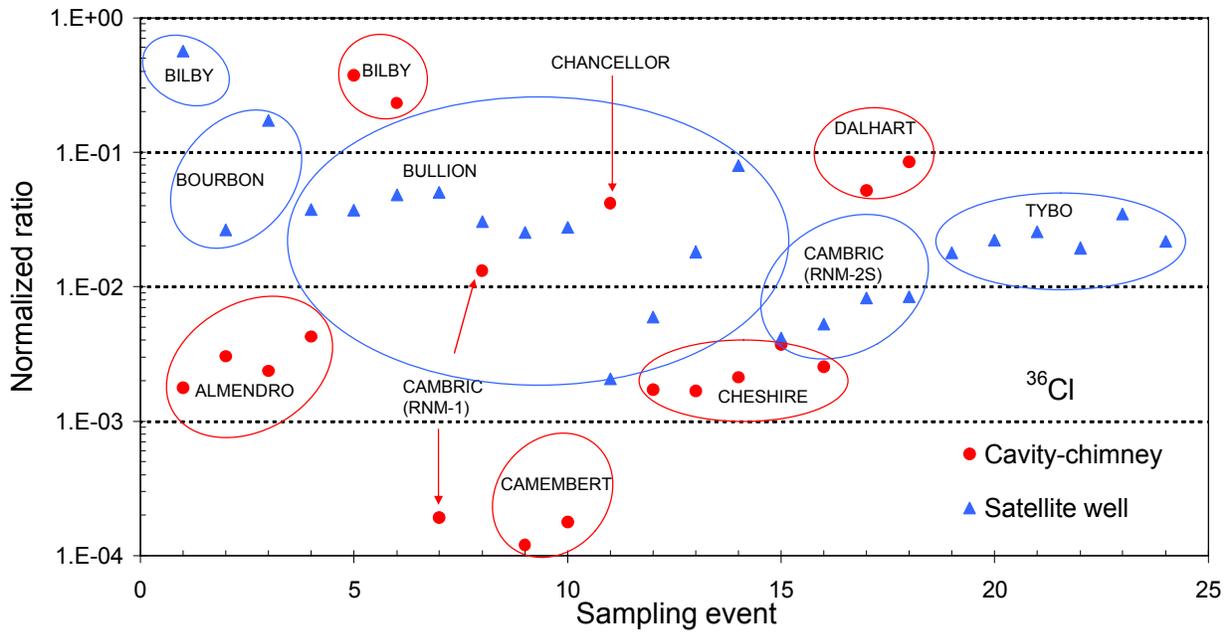


Figure 5

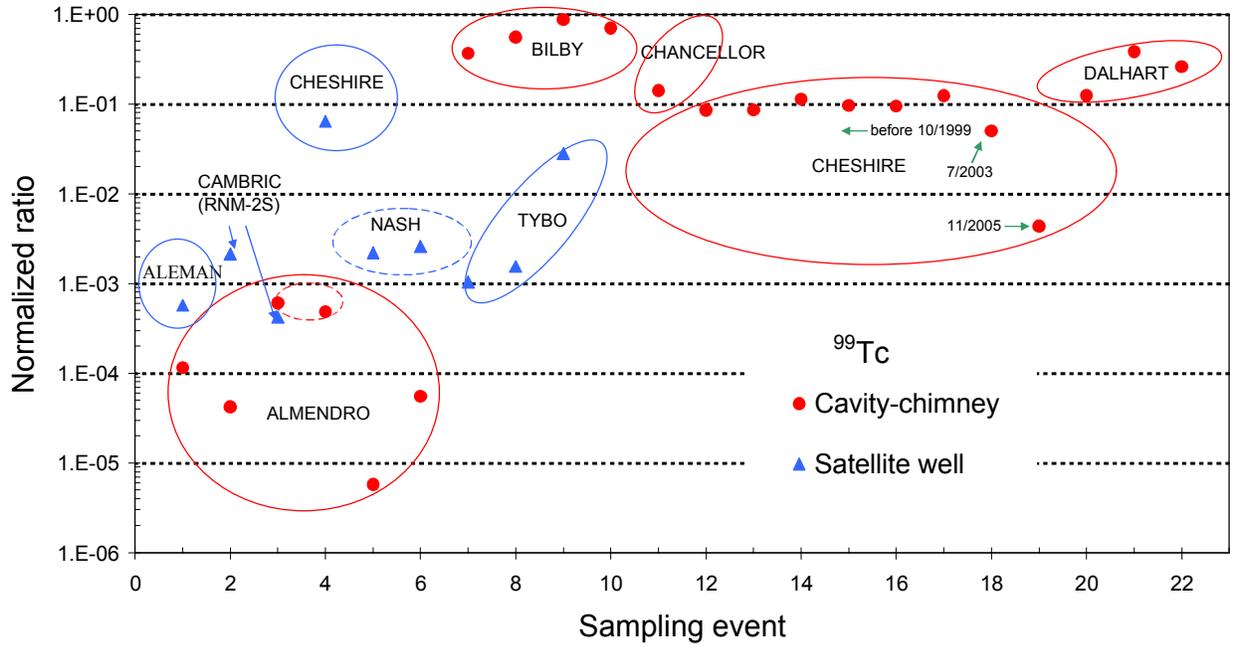
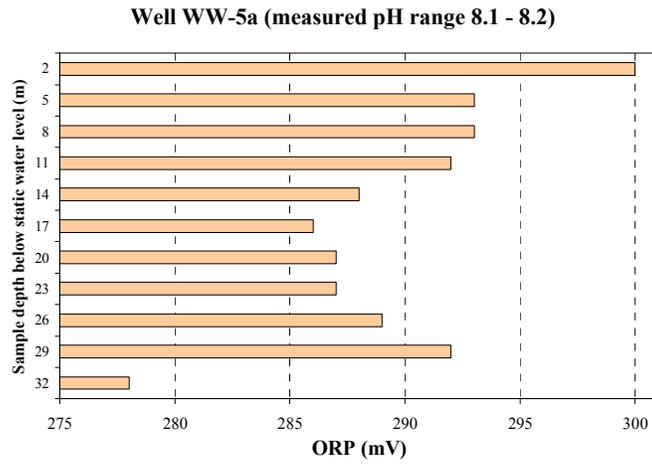
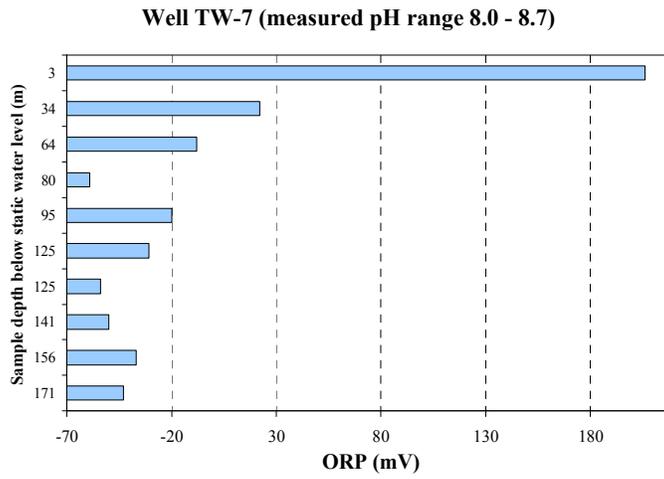


Figure 6
(a)



(b)



(c)

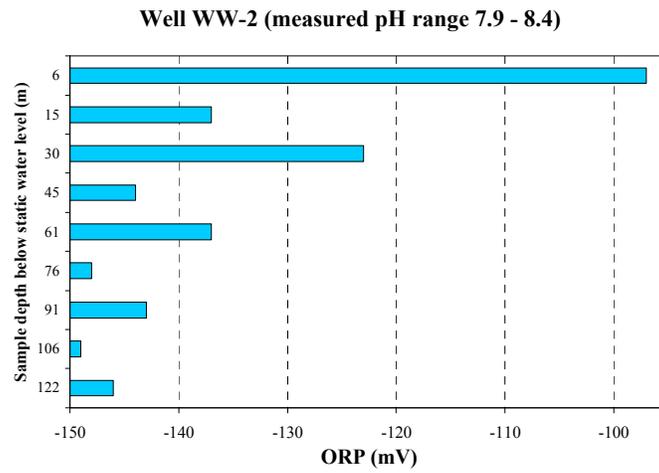


Figure 7

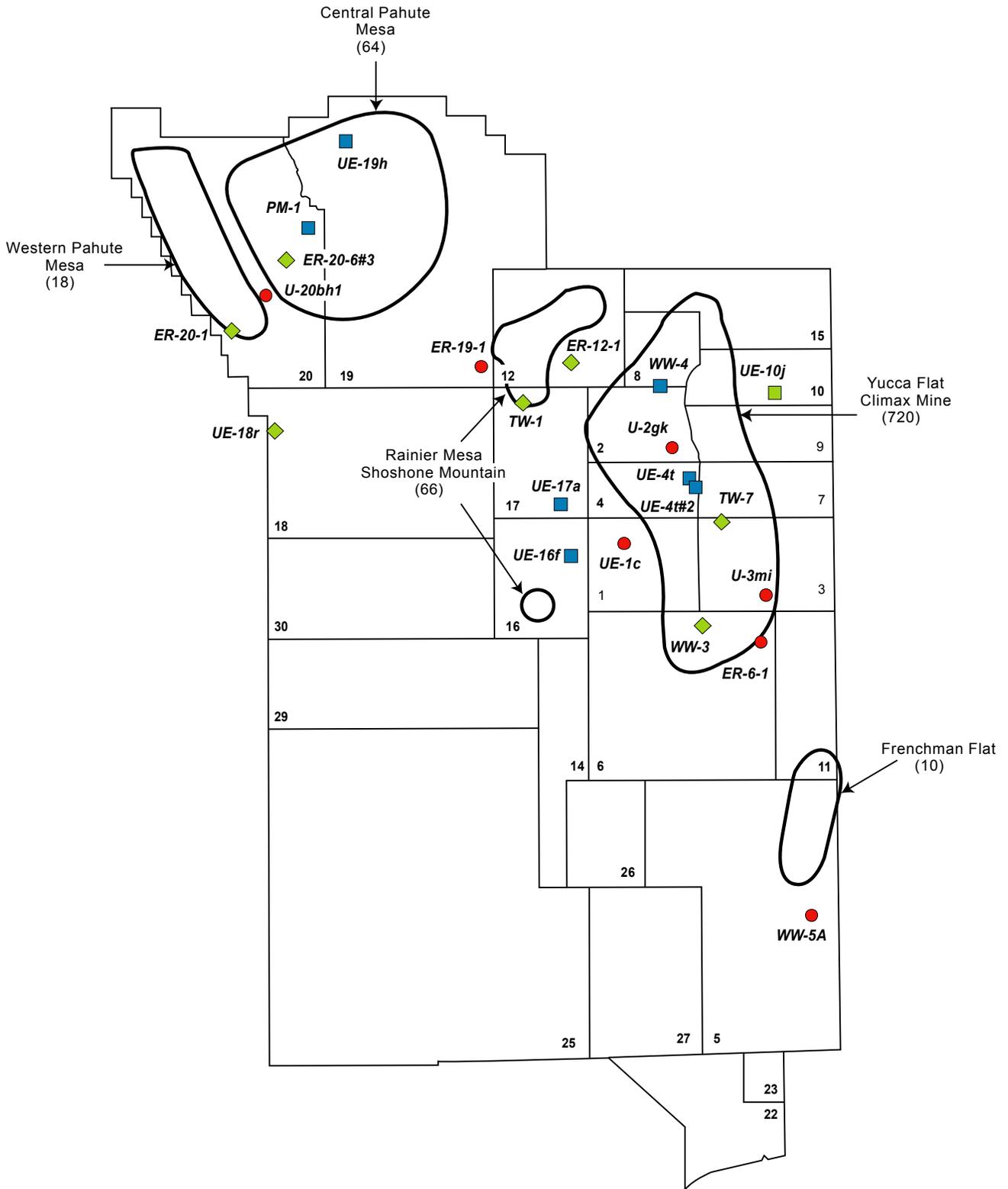


Figure 8

