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Colloid Facilitated Transport of Plutonium at the Nevada Test Site, NV USA

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Abstract Increasing observations of subsurface actinide transport has reinforced the need for a basic geochemical and hydrologic understanding of how these contaminants are transported in groundwater. At the Nevada Test Site, NV, USA, low-levels of plutonium have been shown to migrate on the scale of kilometers. At all but one sampling location the Pu is associated with the colloidal fraction (<1 μm) consisting predominantly of clays and zeolites. The majority of the Pu (∼70%) is associated with the smallest nanoparticle (e.g., colloid) size fraction of 10–100 nm and in one case was identified as associated with a clay colloid. At one unique sampling site, Pu is predominantly associated with dissolved organic matter. Monitoring at over 20 contaminated groundwater sites at the NTS suggests that low-levels of Pu can be mobilized either via colloid-facilitated transport in fractured rock or as a result of aqueous Pu stabilization by anthropogenic levels of dissolved organic matter. Nevertheless, activities above the EPA's Maximum Contaminant Level (MCL) for alpha-emitting radionuclides (15 pCi/L, 0.56 Bq/L) have been measured at only two locations at the NTS.

1 Introduction

Over 800 underground nuclear tests were detonated from 1951 to 1992 at the Nevada Test Site (NTS), as part of the United States' nuclear testing program (DOE 2000) (Fig. 1). Approximately one third of the nuclear tests were detonated at or below the standing water table (Laczniak et al. 1996). An estimated 1.3×10^8 curies (4.8×10^{18} Bq) of radioactivity (calculated to 1992) was deposited and remains in the subsurface at the NTS, consisting of tritium, fission products, actinides, and activation products (Bowen et al. 2001).

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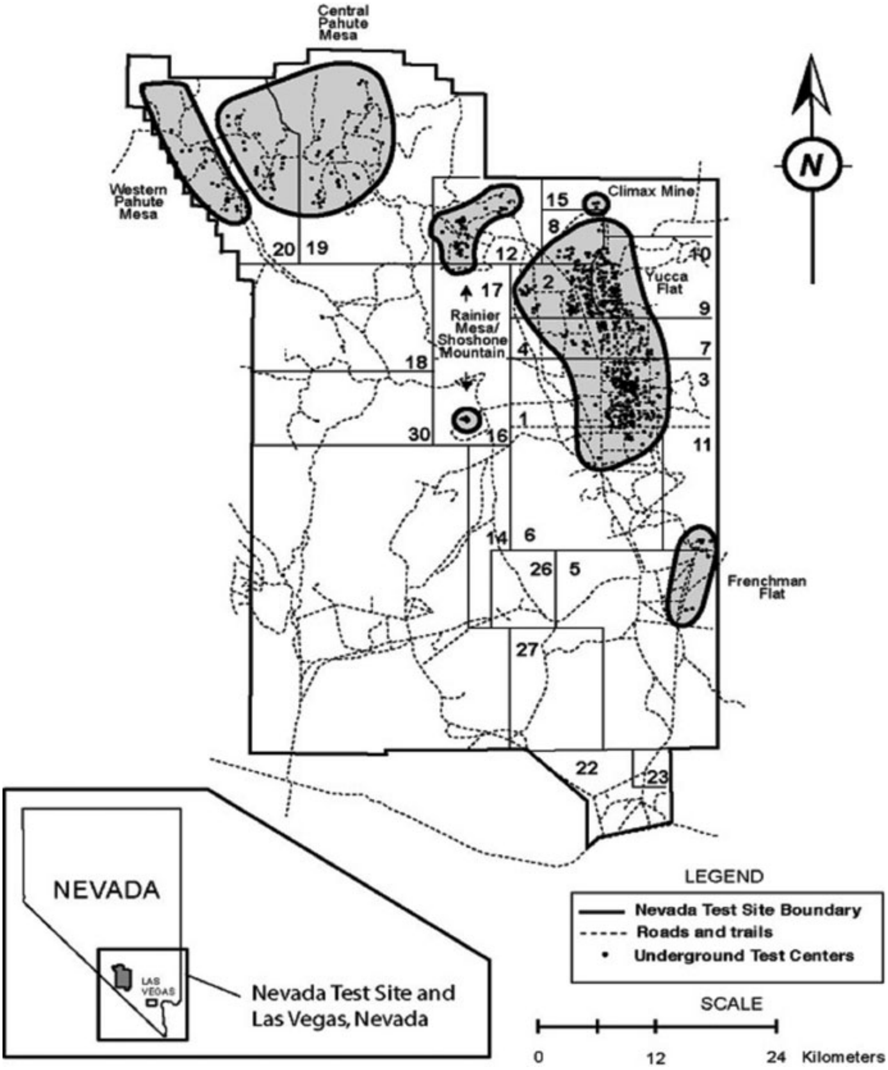

Fig. 1 The NTS showing the location of each of the 828 underground nuclear tests. *Circles* denote geographic regions

The radionuclide inventory at the NTS is similar in magnitude to the inventory at the Hanford Reservation (~1.0×10^{19} Bq) and approximately two orders of magnitude less than estimates for the proposed Yucca Mountain Repository (~1.8×10^{21} Bq). Although the majority of the NTS inventory is currently tritium, approximately 2,775 kg (3.1×10^{16} Bq) is plutonium (Pu) (Bowen et al. 2001). As a result, the NTS contains a large inventory of Pu in the subsurface and thus provides a unique opportunity for studying its fate and transport.

Details of the potential transport of Pu have been a contentious issue especially in the area of nuclear energy and waste disposal. Plutonium, with its low solubility and high surface reactivity, was thought to remain immobile in the subsurface, adsorbed to minerals that constitute the soil and bedrock. Buddemeier and Hunt (1988) implicated colloids in the transport of transition element and lanthanide radionuclides in groundwater at the NTS. In a later field study, Kersting et al. (1999) documented the unexpected appearance of Pu downgradient from its known source and showed that Pu was transported in association with the colloidal fraction consisting of clays and zeolites. Although not a new idea (Kim 1991; McCarthy and Zachara 1989; Santschi and Honeyman 1989; Ryan and Elimelech 1996), colloid-facilitated transport has slowly gained acceptance for the transport of low solubility contaminants. Recent work by Novikov et al. (2006) and Xu et al. (2006) has further advanced our understanding of this process by showing that Pu transported from its source was adsorbed to Fe-oxide and organic colloids, respectively.

These field studies indicate that the Pu associated with mobile colloids can move on the scale of kilometers. Colloidal particles are ubiquitous in both groundwaters and surface waters (Degueldre et al. 1989; McCarthy and Degueldre 1993; Ryan and Elimelech 1996) and are chemically, structurally, and temporally complex organic and inorganic species of both natural and anthropogenic origin. Plutonium can adsorb to colloidal particles, be incorporated within them, or form its own colloidal polymer. Yet, fundamental details of the surface mechanisms that control this process are not well understood and continue to hinder both the conceptualization and predictive capability of colloid-facilitated actinide transport. One of the difficulties is that, in general, Pu measured in field samples is at extremely low levels (e.g., femtomolar Pu concentrations), and this has limited the application of spectroscopic techniques to identify the characteristics of Pu–colloid interactions. Without knowledge of how Pu moves in groundwater, transport models fail to reliably predict the subsurface behavior of Pu. The current inability to effectively predict Pu transport in the subsurface presents a serious scientific challenge.

Information presented in this chapter is based on data accumulated by a number of programs at the NTS that have examined the initial distribution of radionuclides in the subsurface after a nuclear test and its subsequent transport. Most recently, the effort has been led by the Underground Test Area project, Department of Energy, National Nuclear Security Administration, Nevada Operations Office, U.S.A. Details regarding sampling programs and groundwater characterization can be found in Smith (1995), Tompson et al. (2002), and Hu et al. (2008). The focus of this chapter is the observation and characterization of plutonium transport mechanisms at the NTS. We evaluate the Pu detected in several of wells at the NTS, characterize the colloidal fraction of the groundwater, provide a conceptual model for Pu transport at the NTS, and extend this model to other locations.

2 Geology and Hydrology of the NTS

The NTS occupies 2,173 km^2 in south-central Nevada (Fig. 1) and lies within the southern Great Basin region that is characterized by low rainfall and sparsely distributed intermittent streams (Laczniak et al. 1996). The groundwater is deep, roughly more than 250 m below ground surface and flows south–southwest through thick sequences of carbonate and volcanic rocks and discharges at Ash Meadows and Death Valley. Most of the nuclear tests were detonated in either the valley-fill alluvium or volcanic rock. Groundwater near each underground test generally moves laterally through these units and, in some cases, may contact the underlying regional carbonate aquifer. In general, the groundwater in alluvium and volcanic rock is dominantly Na-bicarbonate type with a pH of ~8 and low ionic strength, while the carbonate aquifer groundwater is enriched in Ca relative to the overlying volcanic waters.

3 Phenomenology of Underground Nuclear Tests

During the detonation of a nuclear device, the rock immediately surrounding the device vaporizes and expands as a plasma, temperatures exceed 1 × 10^6 K locally, and pressures exceed 7 Mbar (Fig. 2a). Approximately 70 metric tons of rock is vaporized and another 700 metric tons of rock is melted for every kiloton (kt) of

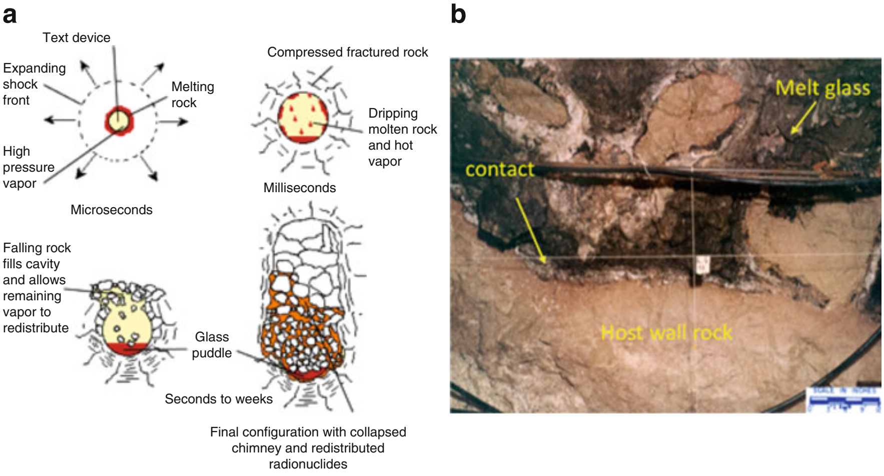

Fig. 2 (**a**) Phenomenology of underground nuclear test, showing nuclear melt glass (puddle glass) coalescing at the bottom of the cavity below the chimney rubble zone. (**b**) Photograph of the Rainier nuclear test (1957) showing the contact between the volcanic host rock below (*pink*) and the nuclear melt glass (*dark brown*) at the bottom of the cavity (Wadman and Richards 1961)

yield (Smith 1993). The majority of refractory radionuclide species [e.g., rare earths (Ce, Sm, Eu), alkaline earths (Mg, Sr, Ba), Zr, and actinides (Am, Pu)] coalesce in the nuclear melt glass at the bottom of the cavity (Fig. 2b). In addition, many of the fission and neutron activation products (^{54}Mn, ^{60}Co, ^{99}Tc, ^{106}Ru, ^{125}Sb, ^{137}Cs, and ^{147}Pm) are also found in the melt glass (Borg 1975). These radionuclides are structurally incorporated into the glass matrix and are not immediately available for transport with groundwater. As the groundwater reinfiltrates the cavity, the melt glass begins to alter, resulting in the slow release of radioactivity into the groundwater. The volatile species (e.g., alkali metals, U, Sb, I, Ru and gases Ar, Kr, Xe) are more broadly distributed in the cavity and overlying rubble chimney created directly above the cavity (Borg et al. 1976). For a more detailed explanation of the nuclear test phenomenology, see: Office of Technology Assessment (1989) and Smith (1995).

Underground nuclear tests ranged in yield from less than 1 kt to detonations in excess of 1,000 kt (DOE 2000) and varied in design and performance. The volume of rock affected, extent of fracturing, thermal perturbations, and most importantly, the residual inventory of radioactivity and its initial distribution in the subsurface are unique to each test. The radionuclides deposited after a test are heterogeneously distributed throughout the cavity, chimney area, and within several cavity radii of the emplacement location. Although the amount of radioactivity at each test may be unique, once the contaminants are deposited, the processes that control their transport in groundwater are similar.

4 Plutonium Detected in NTS Groundwater

Currently, there are approximately 20 wells that can be used to collect contaminated groundwater from either inside or outside a nuclear test cavity (Fig. 3). In addition to high levels of tritium (up to 5×10^7 Bq/L), low-levels of other radionuclides have been detected (Kersting et al. 1999; Hu et al. 2008). Low concentrations of Pu were detected in several of these wells (Table 1), which will be described in some detail below.

5 Well ER-20-5 #1 and #3: Down-Gradient from the Benham Site

The ER-20-5 well cluster consists of two near-field monitoring wells (#1 and #3) located on Pahute Mesa, northwest of the NTS (Fig. 3). These wells were drilled in 1995–1996 to look for down-gradient transport of radionuclides from the Tybo nuclear test. The ER-20-5 well cluster is located 280 m southwest of the Tybo test site and 1.3 km south of the Benham test site (Kersting et al. 1999). The ER-20-5 #1

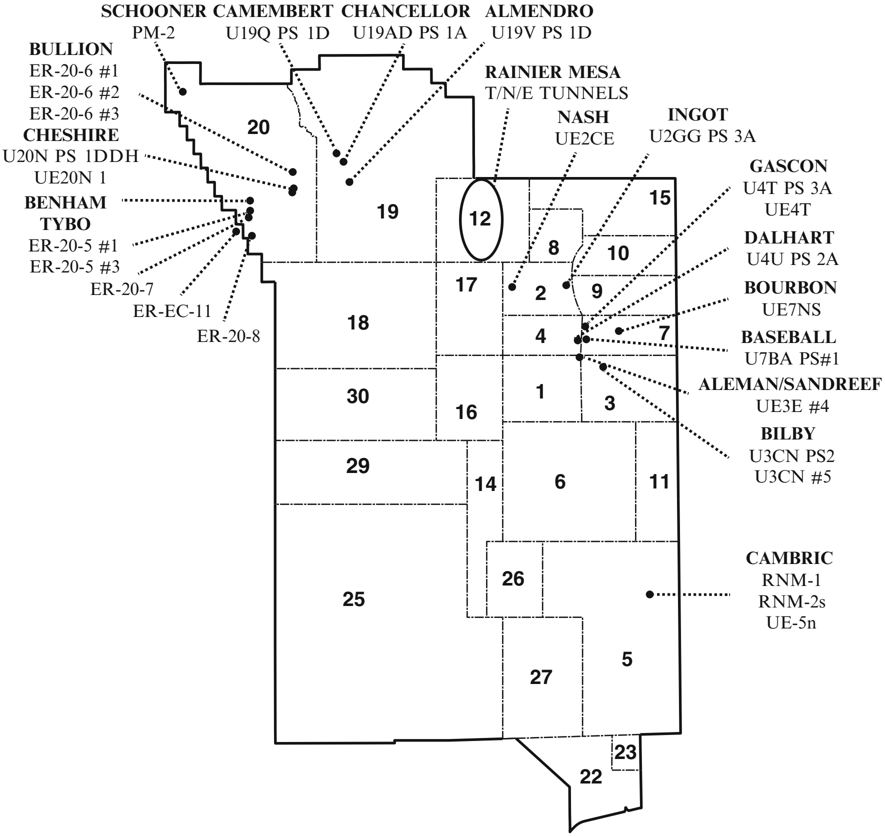

Fig. 3 Map of the NTS showing both the nuclear test locations and the well locations where wells are currently available for groundwater sampling

and #3 wells were screened at a depth of 701–784 m and 1,046–1,183 m, respectively. The Tybo nuclear test was detonated on May 14, 1975, with a yield range of 200–1,000 kt (DOE 2000). The Benham test was detonated on December 19, 1968, with an announced yield of 1.15 Mt.

Groundwater was collected on three different occasions from 1996 through 1997. Plutonium was detected at low concentrations in both wells. In 1997, the ^{239,240}Pu concentration in the ER-20-5 #1 and #3 wells was 0.63 pCi/L (2.3×10^{-2} Bq/L) and 1.13×10^{-2} pCi/L (4.2×10^{-4} Bq/L), respectively (Thompson 1998). Subsequent sampling efforts have measured Pu activity similar to these initial values. The ^{240}Pu/^{239}Pu isotopic ratio of the groundwater from each well was identical and matched that of the Benham nuclear test (Kersting et al. 1999). Despite the close proximity of the ER-20-5 well cluster to the Tybo test, plutonium isotope measurements indicate that the Benham test was the sole source of the Pu activity in both of the ER-20-5 wells.

Table 1 Plutonium concentrations in NTS hot well groundwater samples

Test	Well name	Distance from detonation point, m[b]	Sampling date	^{3}H pCi/L	^{3}H Bq/L	Pu, total pCi/L	Pu, total Bq/L
Bilby	U-3cn PS#2	210 (vert.)	2007	7.7×10^6	2.8×10^5	0.06	2.2×10^{-3}
Camembert	U19q PS1d	462 (vert.)	2003	1.1×10^7	4.1×10^5	<0.02	$<7.4 \times 10^{-4}$
Cambric	RNM-2S	91	2007	1.0×10^5	3.7×10^3	<0.02	$<7.4 \times 10^{-4}$
Cambric	RNM-1	6	2007	8.7×10^2	3.2×10^1	0.006	2.2×10^{-4}
Cambric	UE-5n	600	2004	1.5×10^5	5.6×10^3	<0.02	$<7.4 \times 10^{-4}$
Schooner	PM-2	270 (hor.) 490 (vert.)	2005	2.6×10^5	9.6×10^3	0.006	2.2×10^{-4}
Dalhart	U4u PS2a	140 (vert.)	2008	2.4×10^7	8.9×10^5	0.44	1.6×10^{-2}
Bourbon	UE-7ns	137	2005	1.3×10^2	4.8	<0.04	$<1.5 \times 10^{-3}$
Nash	UE-2ce	183	2008	2.7×10^5	1.0×10^4	<0.008	$<3.0 \times 10^{-4}$
Gascon	U-4t PS3a	54 (vert.)	2008	3.0×10^3	1.1×10^2	<0.0015	$<5.6 \times 10^{-5}$
Gascon	UE-4t #1	168	2008	6.8×10^1	2.5	[a]	
Gascon	UE-4t #2	168	2008	1.7×10^3	6.3×10^1	[a]	
Aleman/Sandreef	UE-3e #4	60/280	2009	1.5×10^6	5.6×10^4	<0.002	$<7.4 \times 10^{-5}$
Tybo/Benham	ER-20-5 #1	280/1,310	1997	6.9×10^7	2.6×10^6	0.63	2.3×10^{-2}
Tybo/Benham	ER-20-5 #3	290/1,340	1997	1.4×10^5	5.3×10^3	1.13×10^{-2}	4.2×10^{-4}
Cheshire	U20n PS1 DDh		2005	3.3×10^7	1.2×10^6	0.46	1.7×10^{-2}
Almendro	U19v PS1ds	125 (vert.)	1999–2006	1.1×10^8	4.1×10^6	<0.004–9.5.	0–0.35
Chancellor	U19ad PS1A	48 (vert.)	2008	1.3×10^7	4.8×10^5	47	1.7
U12n tunnel	U12n extension drift gas sealed plug	450–1,600	2006	1.2×10^6	4.4×10^4	$1.2\ (16)^2$	4.4×10^{-2}
U12n tunnel	U12n main drift gas sealed plug	450–1,600	2006	1.8×10^6	6.7×10^4	$1.1\ (16)^2$	4.1×10^{-2}
U12n.10	Tunnel vent	120	2008	6.3×10^6	2.3×10^5	0.80	3.0×10^{-2}
U12t tunnel	U12t gas sealed door	450–1,000	2006	9.4×10^3	3.5×10^2	$0.009\ (7)^2$	3.3×10^{-4}
U12t tunnel	U12t gas sealed plug	450–1,000	2006	4.8×10^7	1.8×10^6	$54\ (21)^2$	2.0

Note: each well has been sampled more than once, but only one value is listed. For a complete listing of all radionuclides measured at the NTS wells, see Stoller-Navarro (2007)

[a] Not measured

[b] Distance is horizontal unless otherwise noted

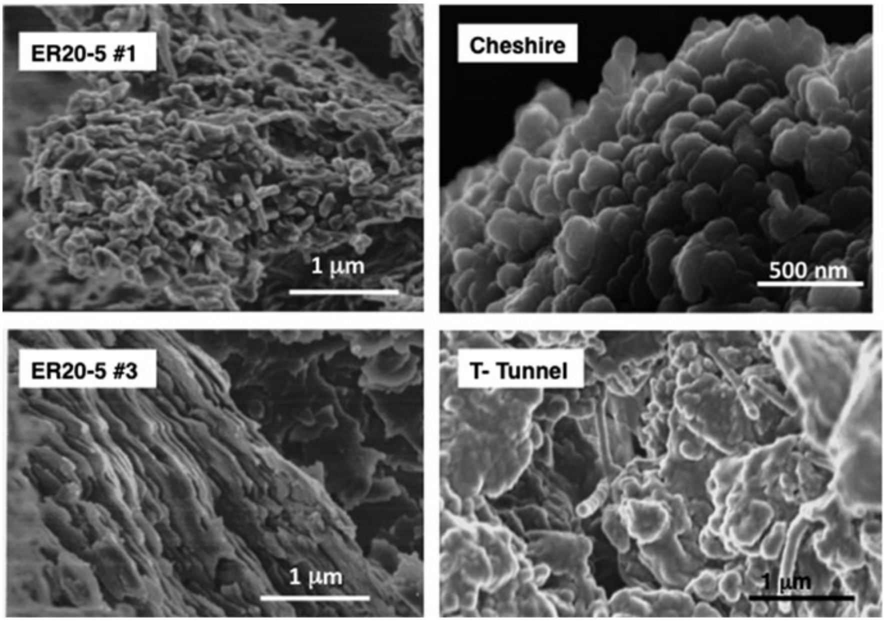

Fig. 4 SEM photomicrographs of colloids filtered from groundwater at the NTS. ER-20-5#1: *rod shaped* colloids are mordenite, ER-20-5#3 platy minerals are stacks of clays

The groundwater from each well was filtered to 7 nm and the colloid fraction analyzed by X-ray diffraction (XRD). The colloidal fraction from each well was the same and composed of clays (illite and smectite), zeolites (mordenite and clinoptilolite), and cristobalite (Fig. 4) (Kersting 2003). There was no difference in mineralogy between the different colloid fractions (>1 µm, 1,000–50 nm, and 50 to ~7 nm). Over 95% of the Pu was associated with the colloidal fraction of the groundwater.

6 Almendro (Well U-19v PS#1ds)

The Almendro nuclear test was conducted on 6 June, 1973, at a vertical depth of 1,063 m beneath the surface of Pahute Mesa. The postshot hole (U-19v PS#1ds) was later developed for use as a near-field monitoring well (Thompson 2001). Groundwater characterization samples were first collected at this site in 1993 at a vertical depth of 942 m; 125 m above the working point. The well was not pumped and sampling was always conducted using a wireline bailer due to the narrow diameter of the borehole. A borehole temperature log run in 1996 recorded a maximum value of 157°C at the bottom of the cavity. The persistence of elevated temperatures more than 30 years after the test suggests that Almendro cavity fluids may be effectively isolated from the surrounding groundwater.

Over the last 10 years, Pu concentrations in groundwater from the Alemendro well have been measured five times, each time producing a different concentration, ranging from nondetect (<0.004 pCi/L) to 9.5 pCi/L ^{239,240}Pu (Rose et al. 2003). The samples were collected 125 m above the working point which is near the edge of the cavity and may not be representative of the average cavity conditions (cavity radius was estimated using the empirical equation reported in Pawloski (1999) with the maximum announced yield from DOE (2000), a depth of burial from DOE (1997), and an approximate overburden density of 2.0 g/cm^3). The samples contained substantial amounts of drilling fluids and as a result, no filtering of the colloids or further characterization was attempted.

7 Dalhart (Well U-4u PS#2)

The Dalhart test (U−4u) was conducted in 1988 with an announced yield of <150 kt (DOE 2000). The working point was at 640 m below ground surface and located in zeolitized tuff. The water level was at a depth of 508 m. In 1990, the postshot drillback U−4u PS#2 was drilled approximately 224 m due south from the working point and at an angle (Mathews et al. 1994; Thompson 1995). The sampling interval is 140–170 m above the working point (2.25–2.75 cavity radii based on estimation methods described in the previous section).

Water samples were collected intermittently at this site since 1992. However, early samples had a significant particulate load resulting from drilling mud (Thompson 1995, 1999). The well was not thoroughly cleaned out until 2003 when 7,058 gallons (26.7 m^3) of water was pumped prior to sampling.

In 2008, samples were collected from Dalhart and the concentration of Pu was 0.44 pCi/L. Groundwater was further separated into colloidal (>40 nm) and aqueous fractions (<40 nm). The colloidal fraction contained approximately 65% of the Pu. It may be surprising that ~35% of the Pu appears to be aqueous (i.e., not associated with colloids). However, it is important to remember that the partitioning between aqueous and colloidal fractions is method-dependent. In this case, the colloidal fraction was only separated to a particle size of >40 nm. Thus, it is quite possible that the colloidal fraction between 10 and 40 nm contributed to the apparently high “aqueous” Pu as noted in the Chancellor groundwater sampling described below. The apparent colloidal Pu fraction should be considered a minimum value based on the size exclusion criterion used.

8 Cheshire (Well U-20n PS1 DDh)

The Cheshire test was detonated on 14 February, 1976, at a depth of 1,167 m beneath the surface of Pahute Mesa with an announced range in yield between 200 and 500 kt (DOE 2000). A postshot hole (U-20n PS1-DDh) was slant-drilled soon after the test and later was converted to a monitoring well for radionuclide migration investigations (Sawyer et al. 1999). The well has been modified several times over its history, but still provides access to the Cheshire cavity and/or chimney region for groundwater sampling.

Groundwater samples were collected in 2005 after the groundwater was pumped from the slant-depth interval 1,250–1,253 m (4,100–4,110 ft) below the surface, within the Cheshire test cavity. Unfiltered groundwater had a total ^{239}Pu + ^{240}Pu activity of 0.46 pCi/L. This result compares very well with earlier Pu measurements and suggests little or no change in the Cheshire cavity/chimney Pu concentrations over time.

Filtration performed on the 1998 and 2003 samples showed that >90% of the plutonium was associated with the colloidal fraction (>20 nm). Colloids were characterized by XRD and consist mostly of clays and minor zeolites (Brachmann and Kersting 2003).

9 Chancellor (Well U-19ad PS#1A)

The Chancellor test was detonated on 1 September, 1983, at a vertical depth of 2,047 ft (624 m) below the surface of Pahute Mesa. It had an announced yield of 143 kt (DOE 2000). The regional water table lies at a depth of 644 m below the surface, located approximately 20 m below the working point of the test (Rose et al. 2008). Groundwater flow in this area occurs principally along interconnected faults and fractures within the thick sequence of Tertiary volcanic rocks that underlie Pahute Mesa (Blankennagel and Weir 1973; Laczniak et al. 1996).

In 2004, a postshot well (U-19ad PS#1A) was drilled in order to retrieve core and groundwater samples in the vicinity of the original test cavity (Rose et al. 2008). Prior to sampling, 69.7 m^3 of groundwater was purged from the well at a rate of 0.09 m^3 min^{-1}. Temperatures were recorded downhole and reached a maximum of 97°C. The high temperatures were observed again in 2008. Persistent high temperatures imply that a significant amount of residual heat remains from the nuclear detonation, even after 25 years. The elevated temperatures appear to have modified the major ion chemistry of the water, resulting in high concentrations of dissolved F^-, Cl^-, and SO_4^{2-} relative to environmental groundwater samples from this same area.

Groundwater collected from the Chancellor postshot well contains one of the highest plutonium concentrations yet observed from a NTS test cavity. The total ^{239,240}Pu activity is 47 pCi/L (~1.74 Bq/L). This is above the Maximum Contaminant Level (MCL) set by the U.S. Environmental Protection Agency for alpha particle emitting radionuclides (15 pCi/L or 0.56 Bq/L). Groundwater was further filtered to determine if the Pu was associated with the colloidal fraction of the groundwater. The colloids were centrifuged and size fractioned at >1,000 nm, 100–1,000 nm and 10–100 nm, and <10 nm (dissolved fraction). Pu was extracted from each size fraction and analyzed using mass spectrometry.

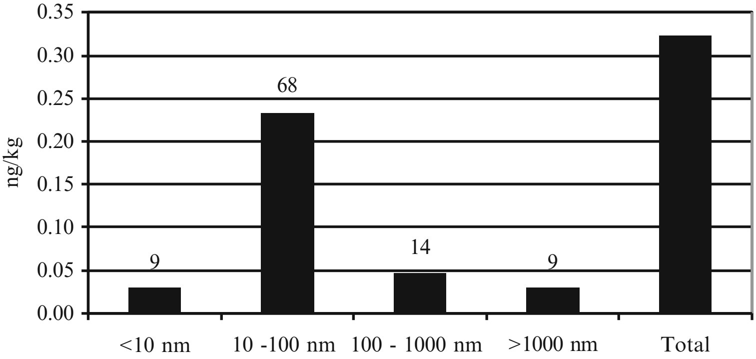

Fig. 5 Plot of total Pu in Chancellor groundwater sample compared to the concentration of Pu in each colloidal size fraction. *Numbers* denote the percentage of Pu in that given size fraction

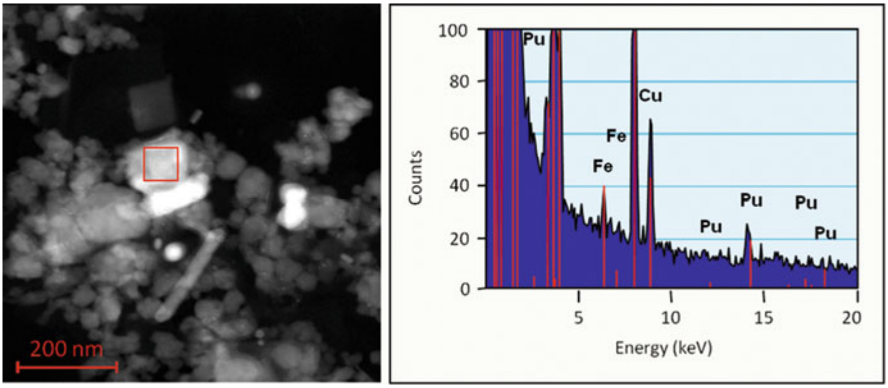

Fig. 6 STEM image (*left*) and EDX spectrum (*right*) of natural colloids from groundwater collected from the Chancellor test cavity. *Box* region shows Pu associated with a clay colloid

Figure 5 shows the total Pu and the distribution in each colloidal fraction. The majority of the Pu (>70%) was in the smallest (10–100 nm) fraction. Less than 10% of the Pu was in the dissolved fraction. Approximately 84% of the total Pu was associated with the colloidal fraction of the water (>20 nm) and when the size cutoff was reduced to 10 nm, the colloidal fraction increased to 91%.

The colloids were further analyzed by XRD and scanning transmission electron microscopy (STEM). All three size fractions consisted of clays, zeolites, and feldspar (Fig. 4). There was no significant difference in the mineralogy between the different fractions. Figure 6 shows a STEM image of Pu associated with a clay colloid and an accompanying EDX analysis of the Pu and clay colloid. Concentrations of Pu in the sample were approximately 10^{-15} M.

10 U12 Tunnel System

A total of 62 nuclear detonations were conducted in the Rainier Mesa area of the NTS, primarily in an extensive and complex system of tunnels drilled into zeolitized tuff of Rainier Mountain (DOE 2000). Activities associated with underground nuclear testing and tunnel construction on Rainier Mesa have altered the tunnel water geochemistry, resulting in much higher dissolved organic matter (DOM) content (15–19 mg C/L) than local spring water (0.2 mg C/L) (Zhao et al. 2011). The source of the DOM is believed to be tunnel lagging, wood debris, drilling fluids, and their microbial decomposition products.

In 2006, groundwater samples were collected from inside two tunnel (U12n and U12t) complexes and analyzed for radionulcides. In contrast to the other groundwater samples collected at the NTS, over 65% of the Pu passed through the 20 nm filter (dissolved fraction). More than 90% of the DOM also remained in solution after filtration. Pu batch sorption experiments were carried out and K_ds calculated for Pu adsorbed to tuff volcanic rock and tunnel fluid with and without the presence of high concentrations of DOM. The K_ds were two orders of magnitude lower when the groundwater contained high concentrations of DOM, suggesting that the Pu is complexed with DOM.

11 Conclusions

At the NTS, where low levels of Pu have been detected in both cavity waters and waters downgradient from the location of underground nuclear tests, the majority of the Pu is associated with the colloidal fraction and not the dissolved fraction. The one exception is when the groundwater has been impacted by anthropogenic increases in DOM, where the majority of the Pu is associated with the dissolved fraction ($>$20 nm). The majority of groundwaters at the NTS are near neutral pH, have low ionic strength and are naturally low in organic matter. When associated with colloids, the majority of Pu is found in the smallest, 10–100 nm colloidal fraction. If filtration cutoff values are larger than 10–20 nm, caution should be used when quantifying the colloidal vs. dissolved fractions as colloidal fraction may only represent a minimum value. At the NTS, the groundwater colloids have a strikingly consistent mineralogy of clays and zeolites with minor cristobalite and feldspar. In groundwater from the Chancellor test, Pu detected with a clay nanoparticle using STEM. Currently there is no evidence that the Pu is transported as its own PuO_2 colloid. The colloidal fraction of the groundwater is able to transport Pu significant distances in the subsurface. Given similar groundwater and hydrologic conditions, colloid-facilitated transport of low-levels of Pu should be expected. This mechanism must be taken into account if reliable transport models are to be developed for Pu and other low-solubility, high reactive contaminants.

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