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YUCCA MOUNTAIN: Earth-Science Issues at a Geologic Repository for High-Level Nuclear Waste

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■ **Abstract** The nation has over 40,000 metric tonnes (MT) of nuclear waste destined for disposal in a geologic repository at Yucca Mountain. In this review, we highlight some of the important geoscience issues associated with the project and place them in the context of the process by which a final decision on Yucca Mountain will be made. The issues include understanding how water could infiltrate the repository, corrode the canisters, dissolve the waste, and transport it to the biosphere during a 10,000-year compliance period in a region, the Basin and Range province, that is known for seismic and volcanic activity. Although the site is considered to be "dry," a considerable amount of water is present as pore waters and as structural water in zeolites. The geochemical environment is oxidizing, and the present repository design will maintain temperatures at greater than 100°C for thousands of years. Geoscientists in this project are challenged to make unprecedented predictions about coupled thermal, hydrologic, mechanical, and geochemical processes governing the future behavior of the repository and to conduct research in a regulatory and legal environment that requires a quantitative analysis of repository performance.

INTRODUCTION

The United States presently has over 40,000 metric tonnes (MT) of spent nuclear fuel (SNF) and 400,000 cubic meters of high-level nuclear waste (HLW) from reprocessing irradiated nuclear fuel (Ewing 1999b, National Research Council 2003a). These high-level wastes are stored in over 100 sites in 42 states. As early as 1955, a committee of the National Academy of Sciences proposed a mined geologic repository (National Research Council 1957) as an appropriate and safe solution for the disposal of nuclear waste. Recently, the National Research Council (NRC) reaffirmed its previous conclusion that "there is a strong worldwide consensus that the best, safest long-term option for dealing HLW is geological isolation." (National Research Council 1990, 2001b). The NRC recommendation for geologic storage is based on the idea that a repository can be built far from human activity in the deep underground and the geologic media itself will isolate the waste from the biosphere. In a geologic repository, the waste could remain safely stored over tens of thousands of years without monitoring or control by society. Despite this consensus, the U.S. government has spent over $7 billion during the past 20 years trying to establish a geologic repository, and the total cost is expected to exceed $50 billion (U.S. Department of Energy 2001). The entire U.S. effort is now focused on a single site, Yucca Mountain, Nevada, and a license application to the Nuclear Regulatory Commission is expected in 2004, with shipment of HLW to commence in 2010.

The quest for a geologic repository has placed the Earth sciences at the center of a scientific challenge that is intricately tied to political and social considerations (Slovic 1987), as well as legal and regulatory constraints. More than any other problem, the selection and evaluation of a site for a geologic repository requires the earth sciences to move from being a "historical" or descriptive science to being a "predictive" science.[1] The present regulatory structure in the United States requires the evaluation of a complex geologic system over a period of 10,000 years. The methodologies for analyzing the predicted performance of a repository originated from approaches developed for the evaluation of nuclear power plant performance (Ewing et al. 1999). In its original form, the analysis was used to assess predominantly engineered systems for which substantial empirical data on reliability existed, with an overlay of Earth-science considerations, primarily in terms of the probabilities of initiating events (e.g., seismic and volcanic activity). Now a wide variety of geologic models (e.g., hydrology, rock mechanics, and geochemistry) are integral to the probabilistic approach to performance assessment for the repository. In this review, we highlight some of the principal geoscience issues that are critical to the results of the performance assessment of the proposed repository at Yucca Mountain.

THE WASTE

The properties of nuclear waste that effect the disposal strategy include radioactivity, chemistry, and thermal output. Although radioactivity is the primary reason for disposing of the waste in a repository, it is the chemical nature of the waste and the heat produced by the waste that have the largest effect on repository performance.

[1]The more recent needs and effort to model global climate change have certainly moved the geosciences firmly into predictive applications. These two efforts, the performance assessment of a geologic repository and predictions of global climate change, share many of the same challenges.

Over 95% of the radioactivity (more than 20 billion Curies[2]) will originate from the used or SNF generated by the 103 nuclear power plants in the United States. A much smaller fraction of the activity will come from vitrified HLW that resulted from reprocessing of irradiated nuclear fuel to reclaim fissile nuclides, mainly ^{239}Pu and ^{235}U. The vitrified waste, a borosilicate glass, still contains the high-activity fission products (nearly 2 billion Curies), such as ^{90}Sr and ^{137}Cs, and will be immobilized in an estimated 20,000 "glass logs," each weighing several tons.

Reactor fuels for light water (LWR) or pressurized water reactors (PBR) typically consist of UO_2 in which the isotopic concentration of fissile ^{235}U is 1 to 4 atomic percent. The radioactivity of SNF is approximately 10^{17} becquerel/MT of fuel largely owing to the presence of 3 to 4 atomic percent of fission products (e.g., ^{129}I, ^{131}I, ^{137}Cs, ^{90}Sr); transuranium elements (e.g., ^{239}Pu, ^{237}Np, ^{241}Am); and the activation products of metals, such as Co, Ni and Nb, in the spent fuel assemblies. The penetrating ionizing radiation (β and γ) comes mainly from the short-lived fission products (^{137}Cs and ^{90}Sr with half-lives of approximately 30 years). The less penetrating radiation from α-decay events comes mainly from the very long-lived actinides (e.g., ^{239}Pu and ^{237}Np with half-lives of 24,100 years and 2.1 million years, respectively). The chemical composition of the spent fuel, when it is initially removed from the reactor, contains hundreds of short-lived radionuclides. With time, the total radioactivity drops quickly, so that at after 10,000 years the total activity is 0.01% of the activity one month after removal from the reactor (Hedin 1997). The total radioactivity of the SNF finally equals that of the radioactivity of the uranium ore mined to create the nuclear fuel after several hundred thousand years (Hedin 1997).

The decay of fission products also generates substantial quantities of heat. The spent fuel generates approximately 2 Megawatts per MT of fuel immediately after removal from the reactor, but the energy release drops to 1300 watts/MT within 40 years. It is important to realize that the first 10,000 years of SNF in the repository are the most complicated because (*a*) the spent fuel still contains substantial quantities of shorter-lived fission products and (*b*) significant amounts of heat are still being generated. After 10,000 years, the thermal output of the SNF is negligible (less than .05% of the original decay heat; approximately 100 watts per MT of fuel) and the radiotoxicity is dominated by a rather limited number of long-lived radionuclides: ^{239}Pu, ^{240}Pu, ^{237}Np, ^{233}U, ^{99}Tc, ^{129}I, ^{79}Se, as well as the remaining ^{238}U and ^{235}U.

THE POLICY FRAMEWORK

The policy framework for the Yucca Mountain project primarily consists of the laws passed by Congress, the environmental standards adopted by the U.S.

[2]Bq = 1 disintegration/s = 2.7×10^{-11} Ci. Or 1 Ci = 3.7×10^{10} disintegrations/s. Bq is the SI unit of radioactivity, although Ci is more commonly used.

Environmental Protection Agency (USEPA) under Congressionally mandated advice from the National Research Council (National Research Council 1995), the regulations promulgated by the U.S. Nuclear Regulatory Commission (USNRC), and the license application by the U.S. Department of Energy (USDOE).

In 1982, Congress passed the Nuclear Waste Policy Act (NWPA) (see Carter 1987 for a history and discussion of the NWPA). The NWPA was a comprehensive effort to outline the U.S. strategy for dealing with nuclear waste and to develop a structure and plan for the responsible federal agencies: the USDOE, USNRC, and the USEPA. The NWPA of 1982:

- Established geologic disposal as the long-term solution for the disposal of nuclear waste resulting from reprocessing of SNF.
- Set the capacity of the first repository at 70,000 MT of SNF (equivalent). A second repository, probably in crystalline rock in the eastern United States, would be used to accommodate the waste above this amount (another 70,000 MT based on the end-of-life projections for presently operating nuclear power plants).
- The USDOE was to initially consider five sites that would be quickly narrowed to three sites for characterization, and then to one final site, which was to be recommended by the Secretary of Energy to the President for licensing. The State with the selected site had the right veto the President's recommendation to proceed, but Congress could override the state's veto by a simple majority of both Houses of Congress.
- A nuclear waste fund was established to pay the full cost of disposal, with generators charged 1 mil/KWh (kilowatt hour) of electricity generated and sold from nuclear power plants.[3] The fund has collected approximately $20 billion from rate-payers (including interest).
- The USDOE was to enter into contracts with the utilities to take possession and begin disposal of SNF in a geologic repository by January 31, 1998.

One of the critical outcomes of this legislation was to create a competition between the sites. A different private sector contractor was responsible for each of the sites. The contractor for the winning site would be guaranteed a steady source of income for many years. Metlay (2000) points out that this created a horse race between the sites, which figured largely in creating a culture of promotion rather than a culture of discovery. The legislation created strong disincentives for defining selection criteria a priori or for determining what might be wrong or unsafe with a particular site.

In 1987, frustrated by the high cost and slow place of the effort to choose a repository site, Congress passed the Nuclear Waste Policy Act Amendment (NWPAA). The amended Act restricted site characterization to the Yucca

[3]A mil is 1/1000 of a dollar.

Mountain, and ended all activities at the other two sites then still being evaluated (Hanford, Washington, and Deaf Smith County, Texas) and deferred indefinitely any activity on a second repository in crystalline rock. Independent oversight of the Yucca Mountain project was strengthened by the creation of the Nuclear Waste Technical Review Board (NWTRB) appointed by the President. The NWTRB is an independent agency of the U.S. Government. Its sole purpose is to provide independent scientific and technical oversight of the U.S. program for management and disposal of high-level radioactive waste and spent nuclear fuel from civilian nuclear power plants.

The State of Nevada and Congress have now exercised the authority delegated to them in the NWPA, and on July 23, 2002, President Bush signed House Joint Resolution 87 approving Yucca Mountain as the designated site. The USDOE is now preparing a license application for review by the USNRC. Although the Act required that Congress submit a license application within three months of the final action, the USDOE plans to submit the application by the end of 2004. The USNRC will then take two to three years to accept or reject the license application. Some twenty lawsuits have been filed by the State of Nevada to challenge the USDOE, the President of the United States, the USEPA, and the USNRC on their compliance with the Federal laws and regulations. The State of Nevada has petitioned the Federal courts to overturn the decision to construct repository at Yucca Mountain.

Regardless of the pros or cons of the Yucca Mountain site itself, the early choice of the site by Congress in 1987, in the opinion of these authors, compromised the scientific integrity of the process. The decision was made in the absence of a USEPA standard or a USNRC regulation for implementing the standard and without completion of many major scientific investigations then underway or scheduled. The USEPA standard and the NRC were not available until 2001 (U.S. EPA 2001; U.S. Nuclear Regulatory Commission 1997, 1999, 2002), and one of the unusual aspects of the standard and regulation is that they are site specific, designed specifically to the geologic attributes of Yucca Mountain. Rather than defining the general safety criteria a priori, and then finding a site post facto that meets these criteria, the USDOE license application must show that the site chosen a priori meets the regulations defined post facto.

Present Situation

The present situation in the United States is that there is a single repository site for HLW. There is no alternative site or strategy, and the entire regulatory structure in the United States is based on the Yucca Mountain site. The present investment in the Yucca Mountain project is over $7 billion dollars, much of which has come from Congressional appropriations. Failure to move forward with the site leaves the United States with no immediate solution for the substantial amount of high-level waste presently awaiting disposal in over 40 states. Some have voiced concern that the HLW is vulnerable to terrorist attack, and much would be gained from the consolidation of these waste at a single site, such as Yucca Mountain. The cost has been high, and progress has been slow. It is within this highly charged political environment that geoscientists study and evaluate the performance and safety of Yucca Mountain. Each scientific controversy, with all of its associated uncertainty, becomes grist for the complicated, often emotional, discussion of this highly controversial project.

THE NATURE OF THE GEOLOGIC CHOICE

In the United States, as in nearly every other country, geologic disposal is the consensus solution among most of the scientific community. Geologic disposal is the strategy defined in the NWPA of 1982, as it is the only technically credible, long-term option that can isolate nuclear waste without continuing active management (National Research Council 2001b, Nuclear Energy Agency 1999). The fundamental concepts behind geologic disposal are that the waste can be isolated in the deep underground and that the geologic medium itself in conjunction with the waste form and other engineered barriers will isolate the waste from the biosphere even in the absence of active monitoring or control by future societies. Geologic disposal relies on the concept of a series of multiple barriers, each designed to limit radionuclide release (Figure 1). These include the natural barriers (the geology) and the engineered barriers (such as the waste form, canisters, and backfill). Processes that must be understood in order to evaluate the performance of these barriers include

- the reactions of the complex radiochemistry of the waste with the cladding, waste package, and backfill material;
- transport paths in groundwater flowing through complex, heterogeneous, faulted and fractured rock;
- interaction of the radionuclides with the geologic media;
- natural hazards, such as earthquakes or volcanic events, that may can affect the integrity of the repository, including its engineered components;
- the effects of climate change; and
- future, inadvertent human intrusion.

The National Research Council was asked in 1978 (National Research Council 1978) to develop geologic criteria for a nuclear waste repository:

- The rock body should be deep enough to isolate the facility from surface processes, large enough to provide a buffer zone to the accessible environment, and be well characterized.
- The repository should be in a structurally stable region in competent rock, and not near a tectonic boundary, active faulting, high geothermal gradients, significant economic resources, potential dam sites, or an area of recent volcanic activity.

- Because of the high uncertainty in predicting the performance of a repository over long periods, a system of multiple barriers (including the geologic barriers of the site itself, as well as the engineered barriers, such as the waste containers, back-fill, and overpack) should provide for a redundant safety margin.
- The probable changes in the hydrologic regime, as judged based on paleohydrologic conditions, should also be favorable.
- Temperatures should be low enough to prevent geochemical and physical reactions that could compromise the repository.
- The rock should provide significant sorption for radioactive materials and not be generally vulnerable to dissolution.
- The waste should be retrievable during the operation phase, but the disposal should be permanent and safe for 10^3 to 10^6 years without supervision.

It is useful to compare this rather sensible set of criteria with the situation at Yucca Mountain:

- The repository horizon is relatively shallow (300 m) in a highly fractured, porous volcanic tuff deposited between 11 and 14 million years ago.
- The repository is in the unsaturated zone approximately 300 m above the water table.
- Geochemical conditions are oxidizing.
- The design temperatures are high, $>100^\circ$C.
- A back-fill (usually a bentonite-based material) will probably not be used, but titanium drip shields will be used to prevent water seepage directly onto waste package surfaces.
- The corrosion resistance of the metal waste package has become an important part of the containment strategy; thus, the long-term performance of the A-22 alloy (56Ni22Cr13Mo3Fe) currently proposed for the containers is a critically important issue.
- The zircaloy (a mainly Zr alloy, 97%, with minor amounts of Sn, Fe, and Cr) cladding on the fuel pellets is presumed to be an important barrier to the release of radionuclides; thus, the short-term integrity of the cladding is important.

The irony of geologic storage is that it is the only identified solution that could be inherently safe and secure in the long-term, but at the same time, the condition of the site at the present time is difficult to characterize, and it is even more difficult to predict the changes that will occur owing to the emplacement of the waste, or at longer times, the changes that will occur as geologic conditions change. As an example, the western United States is a tectonically active part of the Earth crust, and further changes to virtually all aspects of the geologic and hydrogeologic framework are highly probable, even if very difficult to determine in any detail.

Thus, the common sense solution of geologic disposal, with its system of multiple, natural, and engineered barriers, is difficult to implement in a way that is convincing and acceptable to the scientific community and the broader public.

NATURAL BARRIERS

In the HLW program, the term natural barrier refers to the Earth-science processes within the geologic medium that provide protection for the biosphere. In this case, the term barrier is somewhat misleading, as by convention the barrier includes (*a*) the groundwater flow system (which is the main conduit for water to reach the repository, dissolve the waste, and transport the waste to the biosphere), with its potential for dilution and chemical reactions along flow paths, and (*b*) the rock itself, which can act as a sorptive medium to retain the radionuclides in the subsurface. Thus, the geologic barrier is essentially the capability to dilute, retard, and retain radionuclides during transport. In contrast to the engineered barriers, the natural system cannot be designed. It can only be selected or rejected based on site characterization and subsequently predicted behavior.

Geologic Setting

The geologic structure of a site provides the framework for the natural barriers. Yucca Mountain (Figure 2) is located in the South-West part of Nevada about 100 km north of Las Vegas within the boundaries of the Nevada Test Site (a federal reservation where more than 1000 nuclear explosion tests were conducted nearby at the site, above and below ground, between 1952 and 1992). The site is part of the Basin and Range Province. This extensional region is characterized by internal surface water drainage: rivers within the Basin and Range that drain internally into springs, lakes, or playas. Bedrock near the Yucca Mountain project is comprised mainly of bedded volcanic tuffs deposited between 11 million and 14 million years ago overlying Paleozoic carbonates. The rocks are extensively faulted, and there are numerous volcanic features formed over the last 10 million years. The basins between the ranges typically are filled with unconsolidated sediments that may be hundreds to thousands of feet deep.

The tuff is deposited as hot ash, and then fractured during cooling. These are called "cooling fractures" or "cracks." As opposed to through-going faults, cooling cracks are limited to the thickness of each bed, typically a few meters to hundreds of meters thick. Each of the beds differs somewhat in character depending on the details of its deposition and subsequent geologic history. Some are highly consolidated and "welded" by silica during rapid quenching, whereas others are nonwelded, relatively poorly consolidated volcaniclastic sediments. The welded tuffs are more brittle and tend to have more fractures. The nonwelded tuffs are less fractured and contain more zeolites (complex aluminosilicate minerals, known for their ability to sorb metals). Zeolites form over long time periods owing to diagenetic reactions between the original sediments and groundwater. The nonwelded tuff units,

TABLE 1 Hydrogeolgoic units and lithostratigraphy at Yucca Mtn. (after Flint et al. in NRC 2001a)

Group	Unit nomencalture	Lithology
Paintbrush Group	Tiva Canyon Tuff (TCw)	welded
	Yucca Mountain Tuff, Pah Canyon Tuff, and bedded tuffs (PTn)	nonwelded
	Topopoah Spring Tuff (TSw)	welded
Calico Hills Formation (CHn)		nonwelded
Vitric (CHv)		
Zeolitized (Chz)		
Crater Flat Group	Prow Pass Tuff (PPW)	welded
	Bullfrog Tuff (BFW)	welded/nonwelded
	Tram Tuff (TRW)	welded

where present along potential flow paths from a repository to the accessible environment, may form an important barrier to radionuclide transport. Table 1 shows the tuff units at Yucca Mountain. The repository will be in the Topopah Spring unit.

The volcanic tuff contains a significant amount of water. For example, the welded tuffs have approximately 10% porosity, which is nearly saturated with water, and the nonwelded/bedded tuffs typically have 40% porosity and are 25% saturated. In either case, the rock is 10% water by volume (100 liters of water per cubic meter of tuff). Further from the repository horizon, e.g., in the Calico Hills Formation, there are substantial quantities of zeolite. The amount of water contained in the zeolite can be almost as great as that contained in the pore porosity (Flint 1998). Matrix permeability of the welded tuffs is on the order of microdarcies (10^{-18} m^2). For nonwelded tuffs it is somewhat higher (10^{-13} m^2). In general, the secondary permeability, that owing to fractures and faults, is orders-of-magnitude higher than the primary permeability due to matrix porosity.

Yucca Mountain consists of a group of north-south-trending block-faulted ridges that lie to the south of a large Miocene caldera. Some fairly significant faults pass through the repository environs (Figure 3). These tend to be north-south-trending normal faults formed by basin and range extension. The repository site is bounded by the Solitario Canyon Fault to the west and the Bow Ridge fault to the east. The Ghost Dance, Sundance, and Drill Hole wash faults lie within the repository block. Many of these faults in the region are currently deforming at low rates, and some are seismically active (Stepp et al. 2001).

Hydrologic Regime

Yucca Mountain is part of the Death Valley Regional Flow System, which recharges predominantly at various locations to the north of the repository site and discharges in Death Valley. Yucca Mountain is arid to semiarid today, with approximately

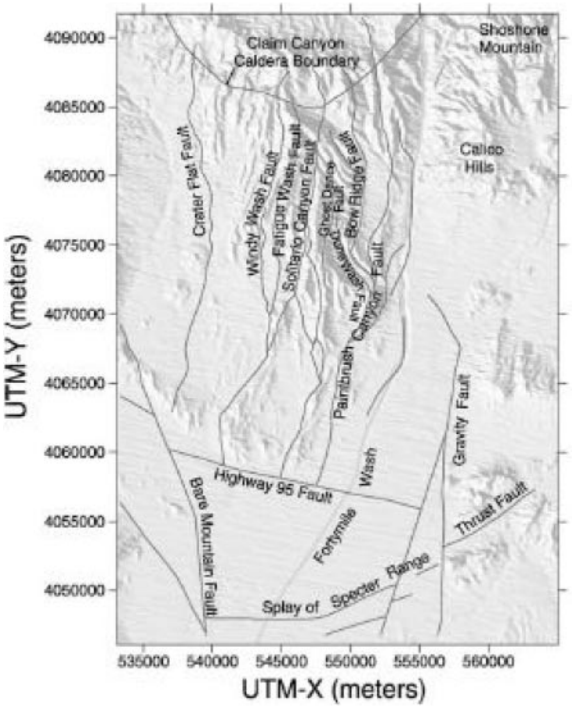

Figure 3 Structural units and tectonic features in the vicinity of Yucca Mountain (from Eddebbarh et al. 2003).

170 mm/year of precipitation directly over the repository block and more than 280 mm/year for higher elevations in the north (Flint et al. 2001). The water table is deep (500–800 m). The hydrologic system is divided into two parts: the unsaturated zone above the water table and the saturated zone below. Regions of saturation can occur above the water table, perched above an impermeable layer. If contamination reaches the water table, it would be transported through deep volcanic and Paleozoic carbonate aquifers. Because the repository is 200 to 300 m above the water table (Dyer & Voegele 2001), the most likely mechanism for water reaching the repository is by infiltration of surface precipitation.[4] The principal natural release mechanism would be a sequence: contact of infiltrating water with waste forms, dissolution of radionuclides, and subsequent transport away from the repository. The important geoscience issues associated with release of the waste are:

[4]Szymanski (1989) postulated that water might also reach the repository by upwelling, possibly associated with an earthquake. This idea was reviewed by a committee of the National Research Council (1992), which found that upwelling probably has not previously occurred and is unlikely to occur in the future.

- How much water can reach the repository over what time period, and where does it come in to the repository?
- How will radionuclide dissolution take place?
- How much water, contaminated with radionuclides at what activities, will travel from the repository horizon to the water table, and how long will this take?
- What amounts of and at what rates will radionuclides be transported in the saturated zone to the biosphere or point of compliance 20 km away?

THE UNSATURATED ZONE The percolation flux is the amount of water that infiltrates at the surface and flows downward (i.e., precipitation that is not lost to evapotranspiration; Flint et al. 2001). The amount of water that reaches the repository is called the seepage flux[5]. Unless some of the percolation flux is diverted laterally away from the repository, the seepage flux is equal to the percolation flux. The original basis for U.S. Geological Survey (USGS) support of a repository above the water table in unconsolidated sediments at the Yucca Mountain site was based on the belief that the percolation flux was so low that the waste canisters would not be exposed to water and corrode; and even if there was corrosion there would be little water to dissolve the waste and transport radionuclides from the repository (Winograd 1974, 1981; Robertson 1982). Consequently, percolation flux is one of the most critical parameters in understanding the movement of radionuclides through the mountain.

Metlay (2000) and Flint et al. (2001) have summarized the history of efforts to estimate the percolation flux at Yucca Mountain. The first estimates of percolation flux were based on expert judgment rather than quantitative analysis. Scott et al. (1983) estimated infiltration at 3% of precipitation and came up with 6 mm/year net infiltration that the authors expected to travel through the fractures. Roseboom (1983) estimated 4 mm/year and, significantly, Roseboom did not characterize the site as dry. Shortly after Yucca Mountain became the only site under investigation, Montazer & Wilson (1984) estimated a percolation flux of approximately 4.5 mm/year, but assumed that the Paintbrush Group tuffs overlying the Topopah Spring repository horizon would act like a "tin roof" (Metlay 2000) over the repository by diverting flow laterally. They postulated that 0.2 mm/year could flow through the Topopah Springs units, and thought that a conservative estimate of the maximum seepage flux through the Topopah unit would be approximately 1 mm/year. Montazar & Wilson largely neglected flux through the fractures because they believed that flow could not occur in the fractures until the

[5]Flux is the volume of water per unit of area at the surface per unit time and thus has the dimensions of length/time (L/T). Flux is the volume of water passing through a unit surface per unit of time; thus, flux is not the same as the velocity of the water, which describes the distance a molecule of water will travel in a given time and thus has the same dimensions, L/T. Flux expresses how much water reaches the repository per unit time, and velocity is how fast it gets there.

rock was nearly saturated. On the basis of Montazer & Wilson's report, the USDOE estimated that there would be zero release of any radionuclide to the accessible environment within the 10,000-year compliance period (U.S. Department of Energy 1984) and the seepage flux through the Topopah would be less than 0.5 mm/year (U.S. Department of Energy 1986). Consequently, the USDOE concluded that the site would meet anticipated release requirements without engineered barriers (U.S. Department of Energy 1986).

From 1988 to 1996, a series of performance assessments were made of the Yucca Mountain site with values of percolation flux as high as 39 mm/year and, in some cases, using models that calculated flow through the fractures rather than restricting flow to the microdarcy permeability matrix. The results of these models showed a strong dependence of calculated release rates on the percolation flux with values much greater than 0.1 mm/year resulting in long-term (post-100,000 years) releases that would exceed the subsequently defined standards by as much as an order of magnitude. In order to evaluate the potential effect of fractures, Gauthier et al. (1992) developed the weeps model, which included only fracture flow. This model accommodated higher infiltration fluxes without causing lateral diversion and calculated seepage levels that would not have met the anticipated USEPA standard (Metlay 2000). Further, Flint & Flint (1995) concluded that the lateral diversion earlier attributed to the Paintbrush tuff would have to be extraordinarily effective to limit seepage into the repository to 0.2 mm/year.

Perhaps the most striking new data was the observation of ^{36}Cl at the repository horizon in the Exploratory Studies Facility (ESF) (Fabryka-Martin et al. 1993). ^{36}Cl (with a half-life of 301,000 years) was produced during atmospheric testing of nuclear weapons in the 1950s and 1960s, and this created bomb pulse increases in $^{36}Cl/Cl$ levels exceeding $200{,}000 \times 10^{-15}$ (Campbell et al. 2003). Elevated values for $^{36}Cl/Cl$ in the subsurface can be used as a measure of the movement of water in the unsaturated zone. Researchers collected samples of rock from the walls of the underground drift (tunnel) in the ESF and analyzed these for $^{36}Cl/Cl$ (Fabryka-Martin et al. 1993; Figure 4). Chlorine is a conservative tracer, as it is not sorbed onto rock surfaces; thus, $^{36}Cl/Cl$ values above 1100×10^{-15} are almost certainly due to the presence of the bomb pulse and indicate water travel times less than 50 years. In locations that correspond to the intersection major faults mapped at the surface with the drift, ^{36}Cl was found to be significantly elevated (Campbell et al. 2003). These data support a conceptual model in which infiltration is controlled by faults and fractures associated with these faults. Faults may control infiltration for two reasons. First, open and connected fractures associated with the faults can form highly conductive channels into the underground. Second, erosion where faults outcrop at the surface may result in topographic lows that collect a disproportionate amount of runoff.

The ^{36}Cl results have led to a number of studies to determine whether rapid transport to the repository horizon occurs. For example, Paces et al. (2003) completed isotopic measurements of samples collected from drill core from boreholes in the drift rather than the drift walls, but detected no elevated levels of ^{36}Cl.

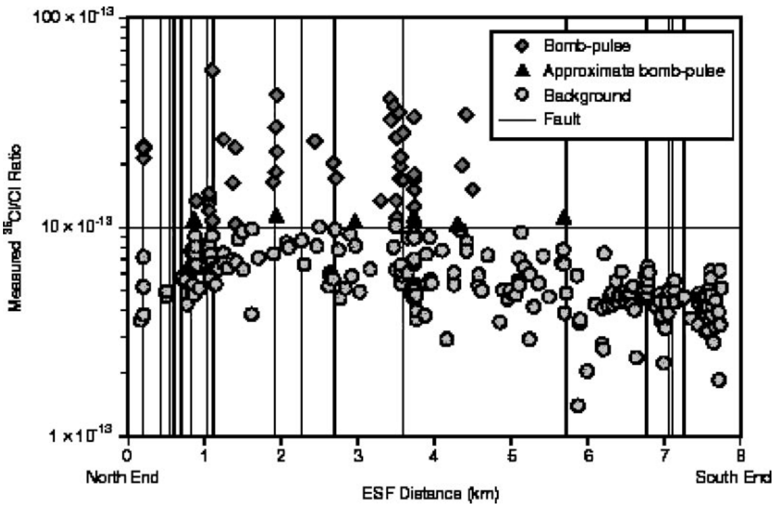

Figure 4 Distribution within the Topopah Springs portion of the ESF of $^{36}Cl/Cl$ ratios and selected structural features (courtesy of J. Fabryka-Martin).

Both the sampling methods and the analysis methods used were different from those of Fabryka-Martin (1993) and Campell et al. (2003). The methods used by Paces et al. (2003) extracted more chlorine from the rock, thus potentially diluting any ^{36}Cl that might have been on the surface of the fractures. For this reason, a peer review panel, convened to examine the different results (Cornett et al. 1998), concluded that “the absence of high $^{36}Cl/Cl$ ratios [in the samples of Paces et al. 2003] does not necessarily indicate that there is no component of bomb pulse water in the sample.”

Several investigators have obtained observational evidence of fast infiltration in laboratory and field studies (Nicholl et al. 1994, Su et al. 1999). Salve et al. (2003) conducted an infiltration experiment in the ESF that demonstrated fast flow along faults. Observations of calcite deposition in the fractures have also indicated considerable fracture flow (Paces et al. 1996). Further, there have been similar observations of anomalously fast flow at other sites (see section below on Performance Assessment and Uncertainty). These all tend to support the faster flow indicated by the reported anomaly in $^{36}Cl/Cl$ by Fabryka-Martin (1993). How can this faster flow be explained?

In porous media and soils, flow in the unsaturated zone is thought to be dominated by capillary forces and gravity (see, for example, Bear 1972, Kirkham & Powers 1972). Capillary forces result from the surface tension developed along a fluid-air boundary that cause a wetting fluid to be drawn into the smaller pores and the nonwetting fluid, air, to be pushed into the larger pores. Essentially, the rock behaves as a sponge soaking up water, with the large pores (fractures) remaining air-filled. For low levels of saturation, only the matrix will conduct water and the fractures will be air-filled barriers to water flow. This is the process assumed by Montazer & Wilson (1984) in their evaluations. As saturation increases, a critical value can be reached where flow commences in the fractures. If capillary forces dominate the behavior of water that infiltrates the ground above the Yucca Mountain repository, then the water would travel through the rock matrix of the nonwelded units, moving downward slowly through the smaller pores rather than the larger, more permeable fractures, which would remain filled with air. Under such a scenario, water should take between 500 and 20,000 years to go from the surface to the repository horizon, and likewise, slowly from the repository to the water table (Flint et al. 2002). Water moving like this through the rock matrix also has much more contact with rock minerals than water moving through fractures. So flow under the capillary model is not only slower, it is more retentive of contaminants.

However, if travel times for water from the surface to the repository are less than 50 years, as implied by the elevated values of $^{36}Cl/Cl$, then it is probably the case that the water that travels this fast is flowing through faults and fractures rather than the rock matrix. In the past few years, researchers have begun to examine the possible mechanisms for vadose zone flow in fractures. Two excellent reviews of this issue can be found in National Research Council (2001a) and the AGU Monograph 42 (Evans et al. 2001). Draglia (1999) has suggested that film flow along the wetted surface of fractures can form waves of water (solitons) that travel quickly downward and could account for the $^{36}Cl/Cl$ travel times reported for the ESF. The solitons carry most of the flux and behave somewhat like rainwater running down a window pane, where drops build up and then course rapidly downward. Tokunaga & Wan (1997) suggest a different mechanism of film flow that depends on a fracture surface transmissivity, which in turn depends on saturation in the matrix. Fabishenko et al. (2000, 2003) suggest pulses of flow may occur owing to dynamical effects having to do with the displacement of air by water in the fracture. Water will pond over the entrapped air until the head builds up enough to displace the air. As infiltrating water fills fractures, it may rise to the point that it can spill over into another intersecting fracture and from there pass downward. The dynamical process results in variable flow rates, much like the classical chaotic behavior observed in a dripping faucet. Even in simple porous media, the invasion of water into an air-filled medium will result in flow fingering and flow concentrating in the fingers where it will move rapidly (Nicholl et al. 1994). The mechanisms described in these papers have been observed on the small scale, but it is still an open question as to how important they are at the site scale. What is clear, however, is that flow in the unsaturated zone is far from uniform and is thus difficult to predict. There is no model that can explain all the observations to date (National Research Council 2001a).

Just because we have evidence of fast flow to the repository, it is not the same as information about the amount of flow (flux) into the repository, and it is the flux that determines how much water will come into contact with the waste packages. At present, there is no accepted conceptual model that explains the travel times and can consequently be used to infer the flux. If climate change were to produce a larger influx of water, saturation in the mountain could increase. Permeability under any proposed model increases nonlinearly with saturation (National Research Council 2001a). Small increases in percolation flux could significantly increase fluid flow through the repository horizon (Nuclear Waste Technical Board 1998, p. 38). This nonlinear response is one of the greatest challenges in predicting the behavior of hydrologic systems over long periods.

THE SATURATED ZONE Given the uncertainty concerning the amount of seepage into the repository, there is more interest in understanding the role of the saturated zone as a barrier to radionuclide transport owing to sorption. At Yucca Mountain, the problem of understanding flow and transport in the saturated zone is complicated by its depth and because it is highly fractured.

The saturated zone consists of two volcanic aquifers separated by aquitards overlying the Paleozoic carbonate aquifer (the Oleana Formation). The groundwater flow pattern in the aquifers is determined by the distribution of permeability, the anisotropy, and the hydraulic head. Heterogeneity and anisotropy are not independent. If the variations in permeability depend on direction, then the heterogeneity gives rise to anisotropy. Such anisotropy is always the case in a bedded rock where the variation across bedding planes is much greater than along the bedding planes.

Throughout the repository region, 150 hydraulic tests in 37 wells were conducted in the saturated zone with only one set of high-quality interference test measurements of permeability available: the C-hole tests (Eddebbarh et al. 2003)[6]. This constitutes a very limited data set describing permeability, and practically no information on anisotropy that have a strong control on the rate, flux and direction of fluid flow.

The easiest of the required parameters to measure is the hydraulic head. The measurement of hydraulic head can be confusing because perched water can be mistaken for the real water table. Because of the paucity of permeability data, the current understanding of fluid flow directions relies mostly on head data (Figure 5) interpreted in light of information on lithological structure (Grauch et al. 1999).

The hydraulic gradients in the region of the repository fall into three categories (Tucci 2001) with low and intermediate gradients to the east/southeast and west of Yucca Mountain and high gradients in the region to the north. Tucci found these high gradients to be 0.06 to 0.07 m/m by contouring the potentiometric surface. Tucci assumed that the measured hydraulic heads of 1020.2 m and 1034.6 m measured in boreholes G-2 and WT#6 in northern Yucca Mountain and Yucca

[6]In a single-hole well test, the same hole is used to withdraw fluid and monitor the response, and the test is consequently most sensitive to conditions near the borehole. In contrast, an interference test has one well to withdraw or inject fluid and another well at some distance to monitor the response. Consequently, the test is sensitive to the entire volume of rock between the boreholes.

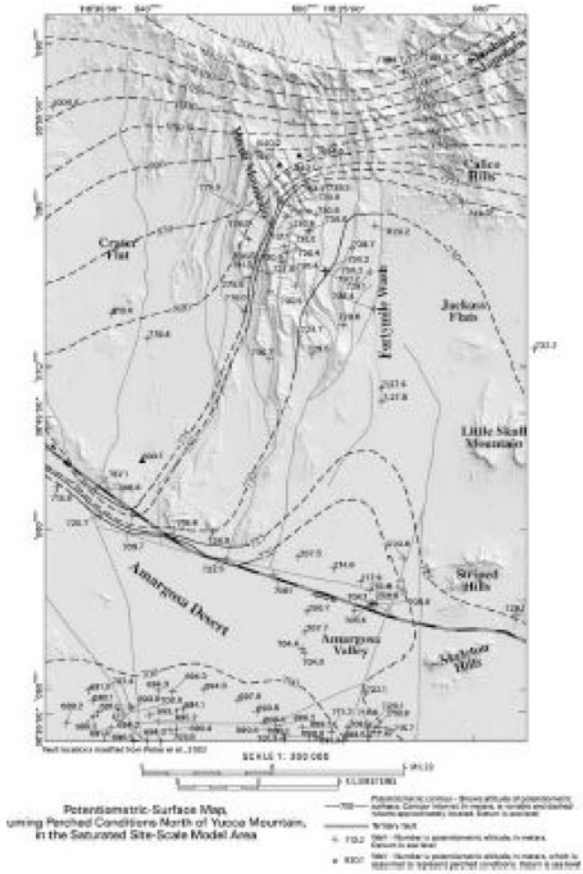

Figure 5 Site-scale potentiometric map and structural features (from Eddebbarh et al. 2003).

Wash represented perched water and did not include these measurements. If these heads are truly the water table, hydraulic gradients would be as much as 0.11. Zyvoloski et al. (2003) studied various alternative models to explain this high gradient because "there is a lack of data to uniquely characterize the Large Hydraulic Gradient." Zyvoloski et al. surmised that the region to the north is altered to a lower permeability by hydrothermal activity from the Claim Canyon Caldera. Tucci (2001) cites several other models to explain the high gradient, including east-west trending faults that contain nontransmissive fault gouge or juxtapose transmissive tuff against nontransmissive tuff, a local stress state that closes fractures, and a highly permeable fault that funnels water directly into the saturated zone. To date, there is no direct observation of such a feature that has a strong effect on both the direction and velocity of groundwater flow.

In the saturated regime, the pattern of open, connected fractures has a strong control on the direction groundwater will flow (Long et al. 1997). Water and contamination flowing in connected, open fractures will be transported along fast flow paths and move farther and faster than in the rest of the medium. It is not easy to characterize the fractures that control flow. Fractures and faults occur on all scales, from the microfractures to major fault systems, and their three-dimensional distribution is difficult to infer in the deep subsurface. Not all of the fractures are permeable, and not all permeable fractures are permeable everywhere along the fracture plane. Some fractures are closed, some are sealed with minerals. Fractures can form or open or close in response to new stresses. The hydraulic conductivity of a fracture is proportional to an effective aperture (i.e., the width, or opening) of the fracture cubed. So, very small changes in opening or closing of the fracture will make large differences in the conductivity. Fracture conductivity is also affected by the amount and distribution of contact between the sides of the fracture, which may change as the fracture opens or closes. Faults can provide barriers to flow as well as dominant flow paths (National Research Council 1996). Because of these characteristics, equivalent porous media models are often poor approximations for fluid flow in fractured rock (Long et al. 1982). It is especially difficult to predict fast preferential flow path behavior with these models.

At Yucca Mountain, nearly all the work done to predict flow and transport uses standard equivalent porous media models to calculate flow (advection) and dispersive transport. For fluid flow, the assumption of an equivalent porous medium, along with the limitations posed by relying mainly on head data, implies that flow is predicted to move in the direction of the hydraulic gradient. However, a calculation of the direction of flow based primarily on hydraulic head could be significantly wrong because flow direction is strongly controlled by the direction of possible flow paths in the open, connected fractures. Unless the fractures are homogeneous, uniformly oriented, and highly interconnected, it may not be possible to make an accurate prediction even of the fluid flow directions, not to mention the rate and flux, by relying solely on the hydraulic head data.

The standard model for subsurface contaminant transport is the advection-dispersion model based on a Fickian representation for dispersion of the waste in the groundwater. For many tracer tests, investigators find that first arrivals are earlier than can be predicted with this model (e.g., Benson et al. 2000). The important aspect of this work is that contamination arrives faster than predicted by the standard model, and consequently, the standard model is not conservative. Alternatives have been proposed that relax the Fickian assumptions and allow for contaminants to be entrained in fast paths. For example, Schumer et al. (2001) and Benson et al. (2000) model dispersion with an infinite variance function, and this leads to an advection-dispersion equation that has a fractional (noninteger) derivative. The equation is easily able to match tracer test data.

The challenge in characterizing the hydrologic system at Yucca Mountain begins with finding that infiltration in the unsaturated zone is apparently more complicated than originally anticipated, and no single model describes all of the field data. Importantly, relatively small changes in conditions or in the conceptual models could result in significant changes in the predicted behavior of the repository. The saturated zone, which was originally thought to be of little importance, has become more significant as a potential barrier. However, the saturated zone will be difficult to characterize because of its depth and complexity, and these studies are not yet informed with a wealth of data. For both the unsaturated and saturated zones, some of the current models may not be conservative.

WATER ROCK INTERACTIONS Although the flow of water determines the direction and distance of transport, chemical interactions determine the mobility of radionuclides. Once radionuclide-bearing waters leave the waste package (see Source Term below), the concentrations are generally modeled as being bounded by a solubility limit. Some of the radionuclides, such as plutonium in oxides and hydroxides, have very low solubilities; thus solution concentrations are expected to be low. Still, estimates are difficult to make because important elements, such as Pu, may exist in several different oxidation states (III, IV, and V for Pu). One approach has been to calculate concentrations based on the formation of $Pu(OH)_4$ as the solubility-limiting phase (Efurd et al. 1998). The initially precipitated solid is probably amorphous, but with time may form crystalline $Pu(OH)_4$. Runde et al. (2002) calculated solubilities and speciation of Pu in low ionic strength waters from Yucca Mountain, and $Pu(OH)_4$ concentrations (in solution as a molecular species) ranged between 10^{2} to 10^{11} M. Concentrations of total Pu are lower by 4 orders of magnitude if PuO_2 is the solubility-controlling phase. Still, there is disagreement over the use of this solubility model because it fails to predict experimental data or the modeled distribution of oxidation states for Pu (Haschke & Bassett 2002).

Once the radionuclide-bearing water leaves the waste package, the main mechanisms of retardation are dilution and sorption. The amount of retardation owing to sorption on mineral surfaces along the flow path is very difficult to estimate. Most estimates are based on laboratory data obtained from batch experiments by which a partition coefficient, K_d, is obtained. The K_d data are only relevant for the conditions of the experiment, and it is difficult to extrapolate these results to the behavior of real geologic systems. More sophisticated column experiments using packed columns or actual core have been used, but these experiments are difficult and time consuming, and results are very dependent on the hydrodynamics of the experiment (Siegle & Bryan 2003) and assumptions that the materials used in the columns are representative of the effective surface chemistry along significant portions of flow paths (which, as discussed above, are not well known spatially). In some cases, field experiments can be conducted using nonsorbing, conservative tracers, but the chemistry of the tracers is, by definition, very different from that of the radionuclides. Field studies of natural analogue sites, such as at the Oklo natural reactors (Jensen & Ewing 2001), have also been used to evaluate radionuclide retardation mechanisms. The most interesting recent results have come from the identification of the solid phases onto which the radionuclides are sorbed, and results have been surprising, e.g., the preferential partitioning of Pu onto Mn-oxides over Fe-oxides (Duff et al. 1999, 2001). Regardless of the approach used to model the change in solution chemistry in contact with the rock, the results are limited by the fact that the experimental, or even field, conditions are not representative of changing conditions along the flow path at a specific site. The frustration in the present effort is that retardation must certainly play an important role during transport along the 20 km to the point of compliance, but convincing estimates of the amount of retardation owing to sorption have proven to be elusive.

COLLOIDS One of the principal barriers to the transport of radionuclides is that they may be sorbed onto mineral surfaces. However, one of the more recent concerns is that sorption onto colloids may actually enhance the transport of radionuclides, particularly those that have a low solubility (e.g., Pu, Am, and Np). There is considerable evidence that the presence of naturally occurring colloids (1 to 1000 nm in size) may enhance contaminant transport (Degueldre et al. 1989, 2000; Kim 1991; Kersting et al. 1999; Siegel & Bryan 2003). There has been recent evidence for transport of plutonium from underground nuclear tests at the Nevada Test Site (NTS) (Kersting et al. 1999). The characteristic ratio of plutonium isotopes for a specific underground test is a fingerprint, which identifies the test that is the source of the plutonium, so the distance and time of transport can be determined—in this case, 1.3 km in 30 years. Greater than 90% of the plutonium was associated with the colloid fraction, consisting mainly of clays, zeolites, and cristobalite. In addition to the colloids that are typically abundant in groundwater, colloids may be generated by the corrosion of spent nuclear fuel and nuclear waste glasses. In the case of the nuclear waste glasses, the waste-form colloids come from surface gel layers where actinides precipitate as they are released during corrosion of the glass.

The challenge is to develop models that capture the effects of colloids on radionuclide transport; however, the type and abundance of colloids depends sensitively on the ionic strength of the solution, pH, and redox conditions (Kersting & Reimus 2003). These are boundary conditions that change during transport and over time. There are very few data on the partitioning of radionuclides onto different types of colloids, and most importantly, the rates of reversible attachment and detachment are not known. Sensitivity analysis of mechanistic models have been used to evaluate the impact of colloid transport on the effective dose (Contardi et al. 2001), but the results of the analysis are very dependent on assumed conditions. As an example, the current USDOE models for colloid transport in the total system performance assessment assume that all colloids from the waste form or those that are naturally occurring can be modeled as either smectite or iron oxyhydroxides; however, Traexler et al. (2003) have found many other phases in the colloid fraction of altered waste forms and in groundwaters from the NTS. Some of the phases, such as monazite, have a high capacity for incorporating actinides. If transport by colloids is an important issue, then much work remains to be done (Kersting & Reimus 2003).

Natural Hazards—Earthquakes and Volcanism

From the perspective of the public, earthquakes and volcanic eruption are the most salient dangers in storing nuclear waste at Yucca Mountain. There are two aspects to each of these natural hazards: What is the likelihood of an event of a given magnitude, and, given this event, what are the likely consequences?

EARTHQUAKES The Yucca Mountain region in the Basin and Range Province is seismically active as it undergoes extension that is accommodated mostly by normal faulting on north-south striking faults. The largest instrumentally recorded earthquake near the repository occurred on June 29, 1992, with a Richter magnitude 5.6, at Little Skull Mountain, some 12 miles from Yucca Mountain. Paleoseismicity evidence exists for regional earthquakes up to about moment magnitude (M_w) 7.5 (Stepp et al. 2001). The significance of earthquakes for the repository is that shaking (ground accelerations) and displacement along faults intersecting the repository may cause structural damage in the repository, particularly to the waste packages.

There are two time periods of interest: the preclosure period, defined as 300 years, and the postclosure period of 10,000 years or more. The preclosure period is roughly equivalent to timescales that are normally considered for other critical structures, such as dams and nuclear power plants that are designed to accommodate probable seismic impacts. The prediction of seismic risk over the postclosure period, i.e., tens of thousands of years, has no precedent. The regulations for the preclosure period focus on determining the magnitude of ground motions that have at least the probability of 10^{-3} of occurring each year during the period. For the postclosure period, the focus is to determine the magnitude of ground motions that exceed a probability of 10^{-4} during the period. For displacements on faults in the repository, the focus is simply to avoid faults that are capable of displacement (Stepp et al. 2001). Assessment of the seismic hazard is called a PSHA (Probabilistic Seismic Hazard Assessment).

A PSHA is conducted in a rigorous manner according to accepted engineering practice. It has three parts. First, the faults that could be seismic sources must be identified and described. Then, an estimate is made of the mean annual rate of occurrence for earthquakes and the magnitude for each source. Finally, an estimate is made of the ground motions that will occur owing to these earthquakes. Each of these estimates has uncertainty that must be evaluated as well. Because the data are incomplete and different experts have different understandings of the processes, the accepted PSHA process uses teams of experts whose estimates are averaged. So the results of the PSHA are strongly dependent on the quality and thoroughness of the expert's efforts.

For the preclosure period, this method probably produces acceptable design data for the repository. It has been possible to compare historically measured ground motions to the predictions as a control on the accuracy of the method. However, for the longer postclosure period, the method has produced rather large values of possible peak ground accelerations (Stepp et al. 2001). The fundamental reason for this is that there is poor knowledge of ground motions close to large earthquakes in extensional geological environments.

Brune et al. (2000) have taken a different approach to this problem by looking at rocks on the surface that are upright, but precariously balanced. By testing or estimating the force that would be required to topple these precarious rocks, Brune et al. can estimate the maximum ground accelerations that could have occurred at the site of the precarious rocks. In other words, if a larger earthquake had occurred, the rocks would have been toppled. The recurrence intervals for these earthquakes can be bounded as well by estimating how long the rocks have been precariously balanced. The results of this approach indicate that the ground motion estimated by the standard PSHA may overestimate the probability of large seismic events by at least an order of magnitude.

The role of earthquakes in changing the hydrologic regime has not been a topic of much study. Clearly, the ^{36}Cl data indicate a strong role for faults in conducting fluids from the surface. Faults that break in an earthquake can create significant new void space in the rock in a number of ways. Extensional openings can form at the edges of the fault or in zones perpendicular to the sense of movement. The fault itself may dilate and, as described above, even small amounts of dilation can create large increases in permeability, which may connect previously isolated parts of the rock mass and provide significant new pathways for fluid flow.

VOLCANISM Volcanic eruptions have occurred in the vicinity of Yucca Mountain since 10 Ma. Eight volcanic eruptions have occurred within the last 1 Ma within 50 km of the site. Three peaks of volcanic activity have occurred within the last 10 Ma, the last one approximately 1 Ma. During the Quaternary, there were three episodes of small-volume, alkali-basaltic volcanism within 50 km of Yucca Mountain (Perry 2003). An eruption that occurred during the operational phase of the repository could disrupt the waste storage system. The questions are: What is the likelihood of such an event, where might it occur, and what would the consequences likely be?

The probability of a future volcanic event is estimated from records of the recurrence of past events. However, Smith et al. (2002) argue that there is a great deal of uncertainty in such estimates and that the mechanisms that support volcanism in the Yucca Mountain vicinity are not well understood. There are several proposed conceptual models for melting and for the relationship of extensional strain to volcanism. Agreement on these models is lacking. Smith et al. (2002) point out that peaks of volcanic activity have occurred about every one or two million years, and, further, we could be at the beginning, middle, or end of a quiescent period; this conceptual uncertainty may be significant.

If there is an eruption near the repository, the effects could include quiet flow of basalt into repository drifts, explosive pyroclastic flow, or the diversion of magma through the repository and subsequent eruption through repository tunnels. Concerns have been expressed that magmatic dykes could intersect the repository or that hot geothermal fluids would circulate through the waste. Interestingly, US-DOE's analysis of radioactive releases during the 10,000-year compliance period are extremely small, but dominated by volcanic events for natural events. Not surprisingly, models that predict the effects of these eruptions are based on many assumptions about the fluidity and amount of the lava, the hydraulic behavior of the objects in the drift, and the possibility for hydrofracture (hydrofracture occurs when fluid pressures in the rock increase to the point that the rock breaks). These are complex phenomena not generally encountered in other engineering projects, and it is difficult to know whether the modeling approach is conservative, much less realistic (Detournay et al. 2003).

ENGINEERED BARRIERS

The engineered barriers are those constructed or engineered by man: the waste form itself, the cladding, canister, backfill, and any other engineered component that is designed to isolate the waste from contact with the groundwater. The behavior of the engineered components are not independent of the natural system. For example, heat from the waste can change the hydrologic regime, or concrete used in sealing the repository can change the geochemical regime. Engineered barriers are designed to isolate the waste from the natural barriers. The major issues are the form of the waste itself, the materials used to make the canisters it is contained in, and the nature of the backfill that may be placed around the canisters.

The most important of the engineered barriers, and the only one we discuss here, is the canister as it is the first line of defense in containing the waste. The design of the canisters is primarily focused on building a container that will not leak over thousands of years. The primary concern is corrosion: How long will it take for the canisters to corrode and the waste to be dissolved or otherwise enter the groundwater?

Because the canisters will be metallic, there are three important elements that contribute to corrosion: water, oxygen and temperature. If you have water and no oxygen, or oxygen and no water, corrosion will be limited. If you have both water and oxygen, corrosion can be a problem. Corrosion is accelerated at higher temperatures. Every other geologic repository program in the world is considering placing waste below the water table. In saturated conditions at depth, the environment quickly becomes anaerobic. Whatever oxygen might reach these depths is quickly used in oxidation reactions with natural minerals and by microorganisms. Without the presence of oxygen, it is possible to find canister materials that do not easily corrode. For example, in Sweden the plan is to use copper canisters (King et al. 2001). If you visit the Kalmar Museum in Sweden, you can see there a copper cannon retrieved from a sunken ship that spent 300 years underwater. The cannon is in nearly perfect condition, thus providing confidence in copper as a canister material in an aqueous, nonoxidizing environment (that was also cold and a well-buffered alkaline solution).

At Yucca Mountain, the original idea was that the repository would remain dry: no water, no corrosion. Even though the repository is above the water table, there are questions about whether the canisters will remain dry. First, past climatic regimes have been much wetter, even in the last 10,000 years, and this could occur again. Wetter climate would increase precipitation and infiltration into the repository. Water could enter the storage tunnels and they could become quite wet, yet not completely filled with water. This means that both air (oxygen) and water would be present. Even in today's climate, some pathways for fast flow of water from infiltration basins to the repository might exist or develop (see Natural Barriers above). The worst case for corrosion is the case where both water and air are present and temperature is elevated. Consequently, the current repository design includes the use of Alloy 22 (56Ni22Cr13Mo3Fe). During its initial exposure to air, Alloy 22 is expected to form a very thin layer of oxidized material at the surface, which will subsequently make the canisters highly resistant to corrosion because it is passive to additional oxidation by air. However, experience with this alloy is only approximately 15 years old. Will it behave in 10,000 years as it does today? How can we know this? Unlike copper under saturated conditions, there is no historical experience with Alloy 22 in unsaturated conditions at high temperatures. Long-term uniform corrosion of passive metals could continue. Although the rate may be small, the timescales of interest are large. In addition, there may be localized corrosion and stress corrosion cracking. Experimental approaches to determining the vulnerability of this canister material to corrosion rely on having realistic boundaries for the environmental conditions, including water chemistry, temperature, radiation, pH, oxidizing potential, ionic species, and microbiological activity (Wong & Payer 2002). The best scientific approach possible is to develop an understanding of the mechanisms of corrosion that may affect this material and to find ways to experimentally speed up time through appropriately increasing the extremes in conditions. Recent results of such studies have prompted the NWTRB to admonish DOE that their assumptions with respect to corrosion of Alloy 22 "lacks a strong technical basis" and

> "Based on its review of data. . .the Board believes that all the conditions necessary to initiate localized corrosion of the waste packages will likely be present during the thermal pulse because of the deliquescence of salts on waste package surfaces, and thus it is likely that deliquescence-induced localized corrosion will be initiated during the thermal pulse. Corrosion experiments indicate that localized corrosion is likely to be initiated if waste package surface temperatures are above 140°C and if concentrated brines, such as would be formed by the deliquescence of calcium and magnesium chloride, are present. Limited data examined to date indicate that dust, which would be present in the proposed tunnels and which would be deposited on waste packages, contains calcium chloride and magnesium chloride salts in amounts sufficient for the development of concentrated brines through deliquescence. (Crevices are widespread on the waste packages, arising from their design

as well as from contacts between the metal and dust particles.)" (NWTRB 2003).

In this highly unusual and strongly worded statement, the NWTRB state urge the USDOE to reconsider the temperature of the repository design (NWTRB 2003).

INTERACTION BETWEEN ENGINEERED AND NATURAL BARRIERS

There are some very significant ways in which the natural and engineered barriers are known to interact, and there may be yet other interactions that have not yet been identified and characterized. The behavior of the waste will change the natural system, and the natural system will consequently change the behavior of the waste package. Problems involving such "coupled behavior" are among the most difficult to solve in the Earth sciences, all the more so because there is no entirely comparable natural system that can be studied in advance.

Source Term

The waste form and its metal container will interact with ground water once water reaches the surface of the waste package and the package is breached due to corrosion. The source term, which is the initial release of radionuclides available for transport, depends on the composition of the water, which will have already reacted with both rock and structural materials, and its flux. If the flux is greater, more water will be in contact with the waste packages, corrosion will proceed more quickly, and the radionuclide release will be greater. The greater the amount of water in contact with the waste, the greater the dilution, but as long as concentrations remain below saturation limits, the waste forms will continue to dissolve and release material.

The composition of the groundwater able to dissolve waste can change dramatically owing to refluxing as the water is driven back into the repository rock during the thermal pulse phase (see Effects of Waste Generated Heat, below). Within the waste package, conditions are difficult to anticipate because of the changing temperature, redox conditions, and the radiation field. As an example, radiolysis of water caused by the ionizing radiation from the spent fuel can lead to even more oxidizing conditions. On the other hand, redox conditions may become more reducing owing to the corrosion of the steel if the oxygen access to the interior of the waste package is limited. The chemical reactions are coupled, and changes in concentration as a function of temperature and pH are nonlinear. Present models use the estimated pH of the solutions in the waste package as the basis for the calculation of the solubility of the radionuclides. Here the uncertainty is high because there is only a limited knowledge of the phases that will control radionuclide solubilities. For example, under reducing conditions the stable solid phase for ^{99}Tc is TcO_2, which is insoluble, but under oxidizing conditions the stable form is TcO_4^-, which is highly soluble (Chen et al. 2000, Campbell 2003).

Initially, all radioactivity is contained in the spent nuclear fuel assemblies (95%) and the HLW borosilicate glass (5%). These waste forms are a barrier to the release of radionuclides. The behavior of the SNF dominates the source term analysis because of its very high radioactivity. The SNF is essentially UO_2 with only 4 atomic percent impurities, which are mainly the fission products (3%) and actinides (1%) created during reactor irradiation.

Although UO_2 fuel was never developed to be a nuclear waste form, under reducing conditions it is a remarkably stable material (Johnson & Shoesmith 1988); hence, most repositories have been placed in sites where the conditions are expected to be and remain reducing. Under oxidizing conditions in the presence of water, or even moisture, the UO_2 in spent nuclear fuel is not stable (Langmuir 1997, Burns & Finch 1999). In oxic solutions, uranium has a strong tendency to exist as U^{6+} in the uranyl molecule, UO_2^{2+}, which readily reacts with a wide variety of inorganic and organic anions to form complexes that increase the solubility of uranium. Throughout most of the natural range of pH, U^{6+} forms strong complexes with oxygen-bearing anions such as CO_3^{2-}, HCO_3^-, SO_3^{2-}, and PO_4^{3-}, which are present in most oxidized stream and subsurface waters. At 25°C and with a typical groundwater p_{CO2} of approximately 10^{-2} atms, the most abundant of these are the uranyl carbonate species, which are stable down to a pH of about 5 (Langmuir 1997). Thus, in most oxic, near-surface environments, uranium is mobile, forming precipitates of uranyl phases that generally have high solubilities (Grenthe et al. 1992). The reaction kinetics for these alteration reactions and the formation of secondary uranyl phases are rapid. Wronkiewicz et al. (1992, 1996) estimated that UO_2 pellets at 90°C in dripping water would have been completely altered in less than 1000 years. This behavior is confirmed by studies of the alteration of natural UO_2 under oxidizing conditions (Finch & Ewing 1992). The alteration can occur simply owing to the exposure of UO_2 in the fuel or to moisture in the unsaturated zone. Secondary U^{6+} phases in natural uranium deposits are typically oxyhydroxides or, depending on the groundwater composition, uranyl silicates or phosphates (Burns et al. 1997c, Burns & Finch 1999). Analogous secondary minerals are expected to form on SNF undergoing incipient oxic-alteration by natural goundwaters at the Yucca Mountain project. The secondary phases may, depending on their structure, incorporate some of the released radionuclides (Burns et al. 1997a,b; Chen et al. 1999, 2000). These secondary phases may finally be the primary source term for the release of radionuclides from the waste package as, by analogy, the secondary minerals from the oxidation of sulfide ores often are the primary source of solute loadings to ground and surface waters at many mine sites.

The Effect of Waste-Generated Heat

As discussed above, the design temperature is a major concern for the repository (U.S. Department of Energy/OCRWM 2002). In addition to corrosion effects, heat produced by the waste will alter the groundwater flow and transport. If the repository design allows for the temperature of the repository to be above boiling, then the behavior of the natural system will be very different than if the waste is cooled prior to emplacement. If waste is emplaced before it is cooled, thermal convection cells may be generated above the waste as steam forms and rises, then cools and falls (refluxing). This type of flow would be entirely induced by the repository and is a permutation on the natural system. Further, rock expands when it is heated, so where the rock temperature increases, the fractures in general will tend to be squeezed shut, causing a decrease in permeability. Hotter fluids dissolve minerals, such as silica, and deposit them again when and where they are cooled down (Dobson et al. 2003). This is commonly observed in geothermal reservoirs where hot silica-rich fluids travel upward, cool, and precipitate minerals that form a seal over the reservoir. Changing temperatures also affect the rates and sometimes even the directions of most geochemical reactions. The net result of all this is a complex coupled phenomenon involving interacting thermal, mechanical, chemical, and hydrologic effects that is very poorly understood. Some have claimed (Buscheck 2003) that a hot repository is inherently safer, as the heat drives away the moisture and thus keeps the waste dry. At the same time, Buscheck et al. (2003) point out that a hot repository may have deleterious effects on corrosion of the canisters, and others point out that a hot repository is so much more complicated that the uncertainty in behavior is too large to accept (Whipple et al. 1999).

The problem is that the amount of waste that has to be stored will cause the repository to be hot unless it is cooled on the surface first or spread out into a larger facility. At this time there is no plan to use interim storage to cool the waste and the USDOE does not plan to make the repository larger. Consequently, these externalities determine a rather major design element of the repository. The NWTRB has pointed out that the USDOE has not yet shown how heat will affect the long-term behavior of the repository and recommend the repository be kept below 100°C (Nuclear Waste Technical Review Board 2002, 2003).

If the waste is emplaced such that there will be a hot repository, there are proposals to cool the repository through ventilation. Calculations show that the repository can be cooled by forced ventilation (fans) or ventilation facilitated by the natural buoyancy of heated air flowing through open access shafts (Danko & Bahrani 2003). These are very interesting concepts, but confirmation of the long-term reliability of this behavior is critical if the safety of the repository depends on cooling.

PERFORMANCE ASSESSMENT AND UNCERTAINTY

The determination by the USNRC that there is a reasonable expectation that the Yucca Mountain site will meet the USEPA standard rests almost entirely on the outcome of a performance assessment (PA). The value of a PA is that it is a structured mathematical analysis that can provide tremendous insight into the connections among the many parts of a complex system. The general features of the approach were developed from risk assessments of nuclear power plants (Rechard 1999). The analysis is based on a systematic identification of all of the natural and

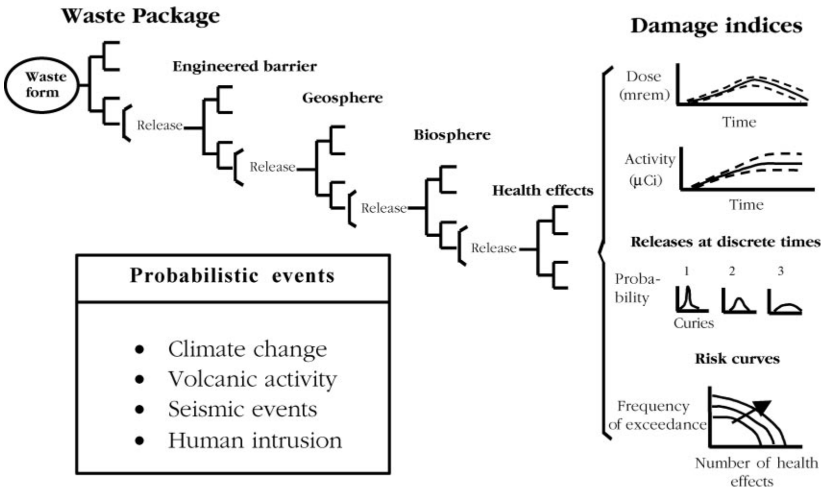

Figure 6 Schematic representation of the steps in a PA of a nuclear waste repository (after Garrick & Kaplan 1994). Depending on the type of regulation, which varies from country to country, the damage indices may vary (e.g., dose in a person/critical population, fractional release of radioactivity over time, or a probability of exceeding a regulatory limit plotted against the estimated effects on health).

anthropogenic features, events, and processes (FEPs) that could significantly contribute to the failure of the repository. All FEPs judged to be of significance are represented by mathematical models that are connected by a single driver code, and the outcome is represented by the projected exposure of an individual or a population at a point of compliance over time (Figure 6). For Yucca Mountain, this is at a distance of 20 km over 10,000 years. The component models are usually abstracted, that is, reduced in complexity in order to meet computational limitations. In a single total system PA (TSPA), there may be hundreds of component models (e.g., geochemical model of waste form corrosion, flow in the saturated and unsaturated zones, climate change, biosphere uptake of radionuclides, etc.) and thousands of input parameters, many represented by parameter distributions, some of which are obtained from expert elicitations rather than any actual measurements. Hundreds to thousands of realizations or simulations are run in order to represent the ranges of expected performance outcomes. There are a number of different philosophical (e.g., deterministic versus probabilistic models) and computational approaches (e.g., Monte Carlo simulations) that may be used. For the Yucca Mountain site, the PA is essentially a probabilistic performance assessment (U.S. Department of Energy 2002) characterized by event trees with probability distributions for model parameters and events. There is a continuing debate over the value and limitations of the PA approach in evaluating the long-term performance of a geologic repository (Ewing et al. 1999, Konikow & Ewing 1999a, Ewing & Macfarlane 2002). Regardless, the USNRC regulation requires the use of PA in determining compliance with the USEPA standard.

During the past decade, there have been a series of iterative performance assessments of the Yucca Mountain site. The two most extensive were the Total System Performance Assessment (TSPA)-viability assessment (U.S. Department of Energy 1998b) and the TSPA-site recommendation assessment (U.S. Department of Energy 2002). A very detailed review of the TSPA (Whipple et al. 1999) provides a good example of the challenges in completing such an analysis. One of the major difficulties is that due to the complexity of the analysis, the inner workings of the component models are nearly opaque to reviewers and the public. A principal concern, most recently expressed by the NWTRB (Nuclear Waste Technical Review Board 2002), is the need to estimate the uncertainty, over time and space, of such an analysis. There are many sources of uncertainty in such an analysis:

- the site characteristics are not fully known
- the range and shape of parameter distributions may be incorrectly estimated
- the probability of the occurrence of important events may not be correct
- there may be numerical errors in the solutions to equations
- assumed boundary conditions may be wrong or change over time.

These uncertainties are further compounded by the fact the component models are highly coupled, and in many cases their behavior is nonlinear, as are the combinations. Small uncertainties propagate through the analysis, potentially causing large and rapid increases in total uncertainty in estimated releases. The PA analysis tries to overcome these difficulties by careful peer review of the component models, by increasing the spread in the parameter distributions, the use of sensitivity analyses to identify the important processes, and by relying on bounding calculations to insure that the final result is conservative or robust with a substantial safety margin. The complexities of the process are such that it remains very difficult to judge how successful these efforts are or even can be.

Despite these efforts to provide a conservative and robust analysis of the behavior of the repository, the underlying assumption is that the analysis actually captures the essential processes in the system. In fact, the greatest source of uncertainty may be the conceptual uncertainty in the description of these important processes. The evaluation of conceptual uncertainty is the most problematic because conceptual uncertainty must include "not knowing what you don't know." A conceptual model is defined as "an evolving hypothesis identifying the important features, processes, and events controlling a phenomenon of consequence (e.g., fluid flow and contaminant transport in the geologic media or corrosion of the canisters) at a specific field site in the context of a recognized problem (e.g., storing nuclear waste)" (after National Research Council 2001a, p. 12). A realistic conceptual model of the behavior of the system depends on having the appropriate understanding of the geometry, physics, biology, and chemistry that control the dominant behavior of the system. If the conceptual understanding is correct, then it is possible to choose appropriate constitutive equations and to define the parameters that govern behavior. However, if the conceptual model is not correct, then the PA analysis may be solving the wrong problem with irrelevant data, and bounding calculations may not bound the actual behavior of the repository. For example, in the case of corrosion of the waste package, if the conceptual model in the PA describes only the interactions of a groundwater in contact with metal, when, in fact, these reactions are facilitated by deliquescence of salts, the results of a bounding analysis may be wrong, and the actual behavior may be outside of the range of the calculated values.

One of the most important phenomena in the analysis of Yucca Mountain is fluid infiltration in fractured rock. There is currently no accepted conceptual model for this phenomenon on the various spatial and timescales required (National Research Council 2001a). Field evidence often does not match predicted behavior. Some rather dramatic examples have emerged where various isotopes or contaminants have been discovered much deeper in the vadose zone than the numerical models had predicted. The ^{36}Cl example described above may be the best known, but there are others. For example, plutonium has been found in the groundwater 200 m under the Radioactive Waste Management Complex overlying fractured basalt at Idaho National Engineering and Environmental Laboratory, whereas previous models had predicted this would not happen for hundreds or thousands of years, if ever (National Research Council 2000). Similarly, the USGS has found anomalous concentrations of tritium at unexpected depths near waste facilities at Beatty, Nevada, where infiltration had been predicted to be zero. Tritium, which apparently could only have come from the waste, was found at depths of 48 m, thus the infiltration was not zero (Prudic 1999). At the Hanford site, transport models predicted that the 90-m-thick unsaturated zone underlying a significant source of radioactive contamination in the “200 Area” would absorb the radionuclides and prevent entry into the groundwater. However, samples from under the leaking tanks revealed ^{137}Cs, ^{99}Tc, and ^{60}Co had migrated much farther than expected, and in some cases, to the water table (General Accounting Office 1989, 1998; U.S. Department of Energy 1999). In these cases, the observations were surprising because they were not and could not reasonably be predicted with the conceptual models that were used. The actual behavior of the geologic system forced the re-examination of the conceptual models.

For even more complex phenomenon, such as coupled mechanical, chemical, heat, and fluid flow in the fractured rock, there is still only a partial understanding of the relevant processes (Whipple et al. 1999). Although data collection has been ongoing for two decades, the NWTRB points out that many parts of the PA (particularly those related to the thermal pulse effects) are based on assumptions, not data, and consequently cannot be relied on in predicting performance or estimating uncertainty (NWTRB 2002). A National Research Council (1990) report points out that the result of two decades of effort at Yucca Mountain is that we now understand that the phenomena involved are much more complicated than expected. Uncertainty has not been diminished, rather the work has “increased the number of ways in which we know that we are uncertain”(National Research Council 1990). To be fair, we must acknowledge that there are uncertainties in every aspect of life, as well as science (Pollack 2003). The fact that there are uncertainties does not mean that we should not take action or move ahead; however, if the uncertainties become so large that they obscure the results of the analysis, then one must take special care in making a final decision.

Conceptual models can never really be validated or proven to be correct (Konikow & Bredehoeft 1992, Oreskes et al. 1994), they can only be disproved. Conceptual models are commonly tested by predicting an outcome and comparing the prediction to a subsequent measurement. This is simple in principle, but confounding in practice because it is often difficult or impossible to predict quantities that can actually be measured and vice versa, or because the act of making the measurement induces changes in the system. Because conceptual models are hypotheses, and these hypotheses may only be disproved, not proved, the most important issue in developing a realistic conceptual model is to collect information that actively challenges the conceptual model. Finally, confidence in a conceptual model does not come entirely from site-specific studies. Rather, confidence comes from the general applicability of the model to a variety of sites, such as natural analogue sites, under different conditions.

The time period of PA is also problematic. The USDOE is required by the USEPA to calculate doses over a 10,000-year period (in other countries the time frame is on a scale of 10^5 to 10^6 years). From the engineering perspective, 10,000 years is a long time to have confidence in the numerical predictions. We have no validated engineering experience in predicting complex phenomena over such timescales. Although the laws of physics and chemistry will not change over the next 10,000 years, we cannot be sure that we have analyzed the right physics and chemistry that will come into play in that period. Although ten thousand years is very long from an engineering perspective, it is quite short from a geologic perspective. When the recurrence interval of major geologic events is on the order of one to ten million years, it is difficult to use a probabilistic understanding of what will happen during periods orders of magnitude shorter. Thus, the probability of geologic events, such as seismic and volcanic activity, are difficult to estimate with confidence within the 10,000-year time frame.

The USNRC will determine whether the TSPA prepared by the USDOE for the license application demonstrates a "reasonable expectation" of compliance with the USEPA standard. At present, a list of hundreds of critical technical issues are already under active discussion between the USNRC and the USDOE, even before the submission of the application in 2004. Despite the effort devoted to resolving these technical issues, this begs the question of whether the method of analysis provides a useful basis for the judgment of the safety of the repository.

ALTERNATIVE STRATEGIES

Yucca Mountain is a complex, one-of-a-kind project conducted in a contentious legal and complex policy environment. The project requires significant scientific advances in many of the disciplines of the geosciences (e.g., site characterization, hydrology, geochemistry, etc.). Perhaps one of the most helpful strategies in this situation would be to prepare a safety case in the sense adopted by nearly all other countries (National Research Council 1999, 2003b) and recommended by the NRC (2003b). A safety case may include a probabilistic PA such as required by U.S. Nuclear Regulatory Commission regulations, but the focus of a safety case is an explanation of the reasons the repository is thought to be safe. It describes in one place the conceptual models that underlie the reasoning, the data that support these conceptual models, the assumptions that have been made and the basis for these assumptions, and the uncertainties that could result from limitations in understanding. The safety case should be quite transparent and include lines of reasoning, rather than thousands of calculations. The safety case should be used to drive decision making in the project (National Research Council 2003b). The NRC (1990) recommends an adaptive management for geotechnical problems where data about the Earth-system is sparse and the system often turns out to be more complex than originally conceived. The recent NRC (2003b) report *One Step at a Time* recommends that iterative review of the safety case is the fulcrum around which programmatic and investigative decisions are made in a step-wise, adaptive management approach. In other words, the implementer of a repository program (in this case the USDOE) uses the description of uncertainties in the safety case to design a new stage of data collection. When the new data are collected, they are analyzed and used to revise the safety case and define a new stage of data collection. In this way, it is the understanding of the underlying safety concepts that drive program direction and constructive learning.

CONCLUSIONS

In this review, we have tried to briefly summarize some of the important geoscience issues that bear directly on the suitability of Yucca Mountain as a nuclear waste repository. Clearly, the scientific issues remain a major challenge but, surprisingly, there are no real technical surprises in our conclusions. A number of the specific issues, e.g., thermal effects on the hydrologic and geochemical systems, were anticipated a quarter of a century ago (Bredehoeft et al. 1978), and Scott et al. (1983) anticipated the role of fractures in the vadose zone. The surprise is that after so much effort, there is so little consensus on even the types and quality of knowledge that are actually required to provide some reasonable expectation that a geologic repository will safely isolate nuclear waste from the environment. To expand the metaphor from C.P. Snow's 1959 lecture, "The Two Cultures," part of the difficulty may be caused by the real and continuing clash among a number of different cultures. In one camp, we have geoscientists who could easily work and argue over the geology of Yucca Mountain for several more decades, and, perhaps, still remain unsatisfied with the state of knowledge. For the geoscientists, the advances in understanding are based on challenging the data and fundamental conceptual models of how the mountain works, but often with limited appreciation for the impact of new knowledge on the main issue—the safety of the repository. In another camp, not too distant in language or culture, we find a community of engineers strongly influenced by their experience with systems analysis and risk assessments that have developed from the early efforts to analyze the performance and safety of nuclear power plants. One sees this influence even in the language. The geologic repository for transuranic waste in New Mexico is a Waste Isolation Pilot Plant (WIPP). To the engineers, models are validated, experiments are confirmed, and decisions are risk-informed.

Of course, these two cultures are not so distant, and they share a common foundation, using the same techniques based on the same fundamental understanding of the science—but there are some important differences. The most important difference is that the temporal and spatial scales that are the common basis for our engineering experience cannot simply be extrapolated to the relevant scales of time and space for a geologic repository. The compliance period of 10,000 years is probably the temporal boundary that separates these two cultures: a time too long for reliable engineering and too short to be fully geologic. There are other cultural differences. The belief underpinning every performance assessment is that highly-coupled, complex systems can be decomposed into simpler approximations, which when reconnected, still provide reliable and useful models of predicted behavior. Despite the limitations of the performance assessment methodology, this does not mean that complex systems cannot be usefully described and understood. The difficulty is that the present regulatory framework drives scientists and engineers toward quantitative, long-term assessments that are outside the scope of current practice and, in fact, cannot be validated. At the same time, scant attention has been paid to the power of a more qualitative analysis that could have actually provided an important basis for site selection and repository design.

Enter the third culture—that of the publics with individual opinions and a full complement of political, social, and public policy organizations. Beyond those most directly affected are the larger publics using energy (nearly 20% of the electricity in the United States is generated by nuclear power plants) and paying taxes into the Nuclear Waste Fund (approximately twenty billion dollars) is certainly affected. The common, often stated, wisdom is that "... the biggest challenges to waste disposition are societal" (National Research Council 2001b); but is this true? Certainly, our political structure has failed to produce the solutions for nuclear waste envisioned in of the NWPA of 1982, and much of the present emphasis in the nuclear community is devoted to educating the public and communicating risk. There is a general sense that if only the public understood, we could move forward. Perhaps, the situation is rather the opposite, the public does understand the scale of the challenge. Perhaps, the struggle for public acceptance is a struggle to develop a useful level of scientific understanding. Perhaps scientists and engineers have simply failed to directly and convincingly answer the following question: With what confidence and by what methods can we predict the behavior of a complex geologic system over tens to hundreds of thousands of years? Or more importantly: Is it safe?

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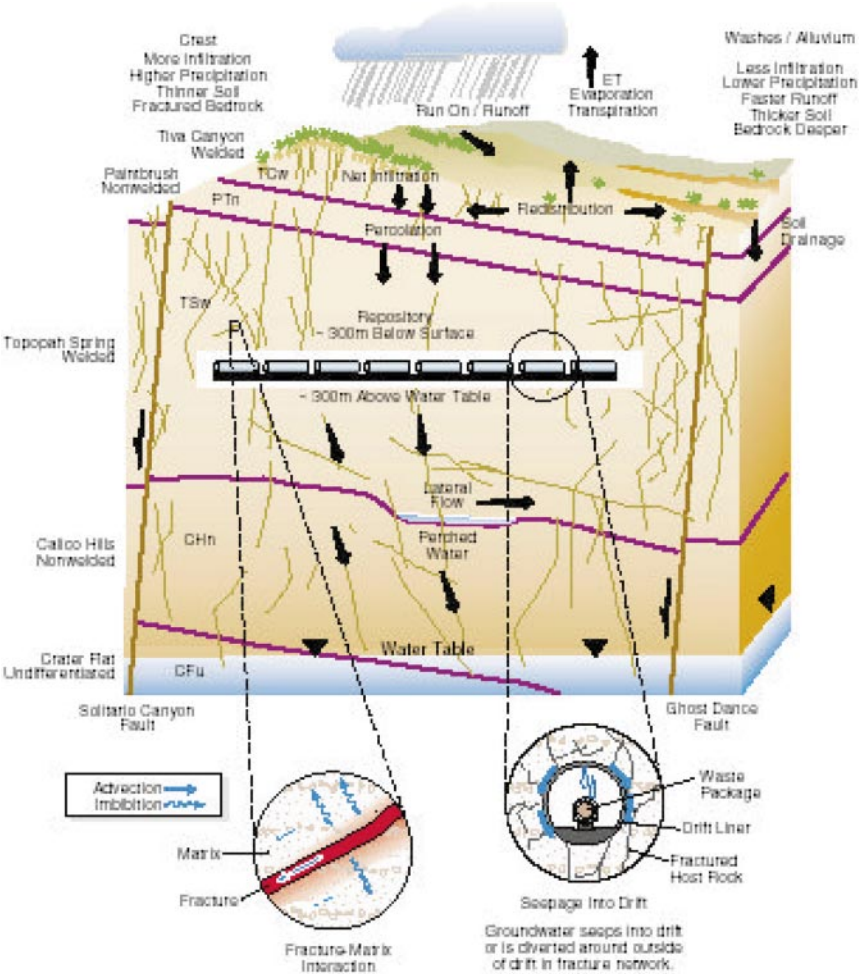

Figure 1 Block diagram showing stratigraphic and hydrogeologic setting of proposed unsaturated-zone (above water table) repository at Yucca Mountain. From Viability Assessment (vol. 3, figure 3–1).

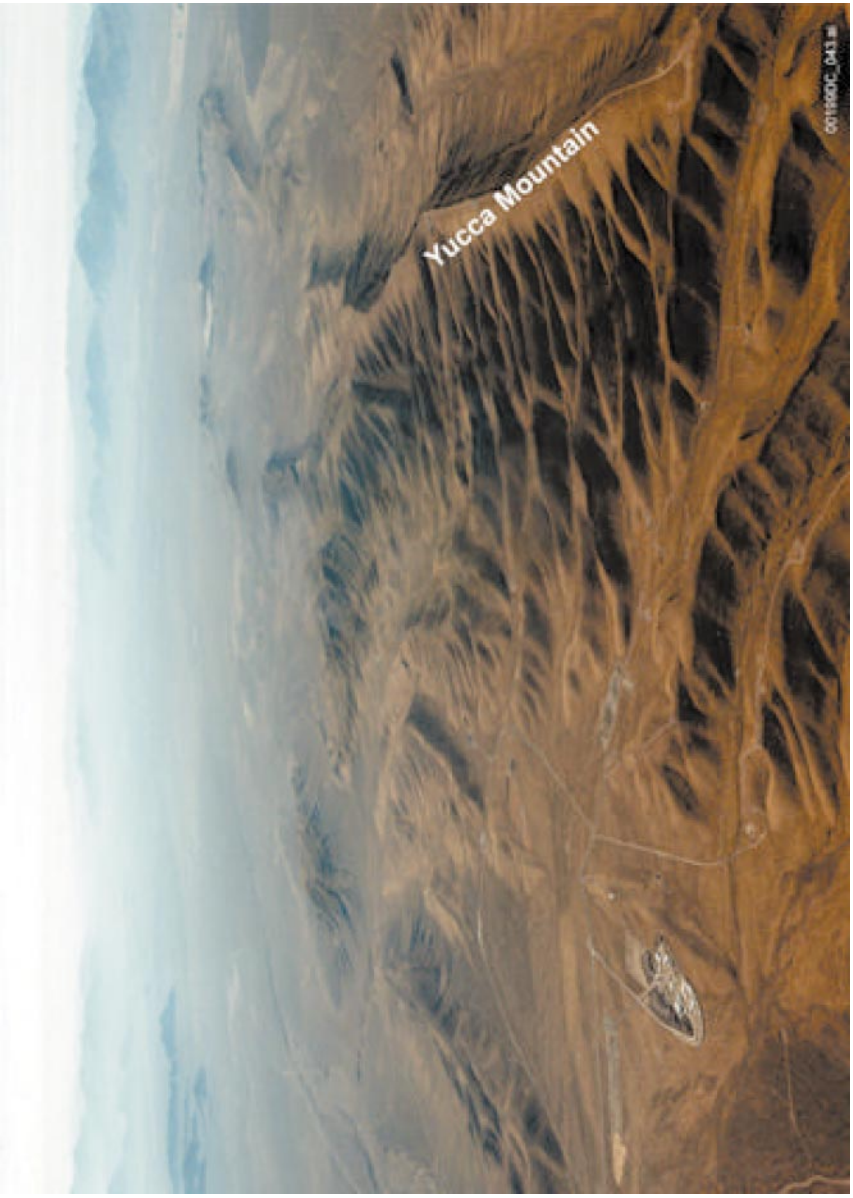

Figure 2 Aerial view of Yucca Mountain looking south. Photograph courtesy of the USDOE.