

The Use of Environmental Tracers as Indicators of Paleoclimate and the Paleohydrologic Response: A Thesis Proposal

NSF Project Title: Development of Environmental Tracers for Water and Solute Transport in Arid Vadose Zones with Applications to Paleohydrology

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Introduction

The unsaturated zones of desert regions have increasingly been called upon as the final resting-place for hazardous waste, both nuclear and chemical. The low precipitation and high evapotranspiration rates result in low recharge potentials. This has led to increased interest in the interaction between water from precipitation, the water table, and the proposed waste disposal sites at depth. Thick alluvial sequences and deep water tables characterize many arid regions. These thick unsaturated zones can often be archives for paleoclimatological information. These archives, preserved in the form of precipitation residues or salts, are readily preserved due to low water contents and even lower hydraulic conductivity. The recharge history of a vadose zone profile can be traced back to the last time significant recharge occurred at the site. This research seeks to tap into this resource for reconstruction of past climate regimes and for further insight into flow and transport processes during these periods. This has direct application to the proposed siting of waste facilities in these regions. Despite extensive research, the transport properties and general response of arid vadose zones to wetter climate regimes are still not well understood.

While the analysis of chloride in vadose zone profiles has been widespread, it has been limited to mostly shallow cores (Dettinger, 1989; Phillips, 1994). In this study, we seek to apply this technique to a previously unanalyzed deep core from the Nevada Test Site (NTS). This deep core, UE-6e, was drilled in 1973, and has been archived at the USGS Core Library in Mercury, NV. With current precipitation rates of less than 15cm/yr at the site, it is hoped that the core may contain in excess of 100,000 years of recharge history. This record may provide insight into climate changes back through the penultimate glaciation. My work, in a joint project with colleagues at New Mexico Tech, will also look into mechanisms for calibrating the chloride deposition rates through time, which is one of the greatest uncertainties in the chloride mass balance method. We will also be looking at stable isotopic signatures as a paleothermometer of soil water. I also plan to extend the analysis of vadose zone profiles to other tracers, not previously developed, such as nitrogen compounds. The development of new tracer techniques will not only have application to this core, but to vadose zone profiles in many arid regions. Following I layout the proposed study plan and schedule for this research.

Research Objectives

1. Using existing data from a wide variety of field sites, and new data using $^{36}\text{Cl}/\text{Cl}^-$ ratios, refine the current and paleoflux estimates of chloride to the land surface.
2. Investigate spatial heterogeneity of Cl^- accumulation in the vadose zone to quantify the possible extent of preferential flow.
3. Using soil water tracers from new and existing core data, develop a record of recharge and paleorecharge in the Yucca Flat area. This data set will be compared to the existing paleoclimate data set from Frenchman Flat and other regional records.
4. Investigate the use of alternative tracers (specifically NO_3) as recharge estimators and develop NO_3 profiles in UE-6e and UE-6s

Background

The development of sound techniques for the description of liquid and solute flux in the vadose zone has been driven by the extensive use of arid regions for radioactive and chemical waste disposal. The low precipitation and high evapotranspiration rates result in low recharge potentials. Unsaturated zone processes have been investigated with both direct physical measurements, such as lysimetry, indirect physical measurement, such as Darcy flux or water balance estimations, or by proxy methods such as the use of tracers. There is general agreement that the indirect physical measurements were the least successful and the use of environmental tracers was the most successful (Allison, 1994). The use of environmental or natural tracers, using the chloride mass balance (CMB) method, is one of the cheapest and most useful methods for determining recharge rates. In this method, a mass balance approach is used with the chloride ion. Precipitation and dryfall are the inputs to the top of the system. Differences between soil water chloride and precipitation chloride are attributed to evaporative flux. In arid regions with low recharge, the tracers present in the vadose zone can represent tens of thousands of years of recharge history.

The only direct method of measuring moisture flux through the unsaturated zone is the use of lysimeters. These can be very useful for water balance calculations, measurement of evaporation and in some cases to quantify deep drainage. The downsides of using lysimeters are that they are difficult to install and expensive to maintain. The indirect physical measurements rely primarily on two methods, the use of Darcy's law to measure fluxes and the use of the water balance approach to account for total flux across the system. A simple expression of the water balance is,

$$\mathbf{R = P - E_a + S}$$

R is the recharge term, **P** is the total precipitation, **E_a** is the actual evapotranspiration and **S** is the change in storage. The main difficulty in this method is the determination of evapotranspiration. In regions where evaporation and/or transpiration are high, downward fluxes are such a small percentage of the water budget that the uncertainties in the measurements are much larger than the flux to be measured.

Another method of indirect measurement is to quantify the water flux below the root zone. Measurement of the water content below the root zone and subsequent estimation of the transmissive properties of the soil. If the governing equation for the Darcy flux is given by,

$$\mathbf{R} = -\mathbf{K}(\theta) \nabla H_t$$

Where **R** is the recharge (water flux), **K** is the unsaturated hydraulic conductivity, which is a function of water content (θ), and ∇H_t is the gradient in hydraulic head. The hydraulic conductivity as a function of water content is difficult to measure in the field and you are again crippled by the fact that very small errors in water content translates into very large errors in unsaturated hydraulic conductivity. Values for cumulative flux have been shown to vary by a factor of five (7-37 mm/yr) depending on the method used to calculate unsaturated hydraulic conductivity (Allison, 1994)

Both the direct (mass balance) and the indirect (Darcy) methods for estimating recharge suffer the problem of needing a long record to estimate yearly or decadal fluxes. Temporal variability in arid regions is large enough that it is difficult to obtain a significant record of measurements to come up with a representative value. Additionally, unless a large number of samples are taken, the physical methods do little to measure the spatial variability of recharge based on varying surface topography and changes in soil type.

Tracers

The use of natural tracers to estimate recharge has several advantages over the physical methods. The recharge record represented by the tracers often represents a historical record, much longer than any record available by physically taking samples. A historical record can only be obtained if a reasonable history of precipitation and solute flux to the land surface is known. Principle tracers used in these studies are as follows; ^3H , ^{14}C , ^{36}Cl , ^{15}N , ^{18}O , ^2H , ^{13}C , and Cl. The first three are radioactive and their natural abundance has been significantly modified by above ground testing of nuclear weapons. These high concentrations of "bomb pulse" radionuclides are generally found in the very near surface. Bomb pulse tracers are not expected to be important in the deep core we plan to study.

The ideal tracers are those that are actually present in the water molecule, isotopes of hydrogen or oxygen. Chloride and nitrogen are considered to be conservative, that is, to move with the water, in most soils. Some soils may exhibit an anion exclusion property, which will cause the ion to move ahead of the water being traced. While nitrogen is not truly conservative, because it

is taken up by plants, it can be considered to be conservative beneath the root zone or in areas of low biologic activity (Edmunds, 1997). Carbon is by no means conservative, so to use it as a tracer, all sources and sinks of carbon in the system must be accounted for.

There are basically three techniques for estimating recharge from tracer profiles. Different techniques are applicable to different isotopes. The first is the estimation of the age from the tracer peak. If the time of a peak production of a given isotope is known, that spike can be seen in the subsurface and the travel time determined from the surface to the point in question. This is most applicable to radiotracers released during weapons testing. The second is the shape of the tracer concentration with depth in the profile. In this method the tracer is examined throughout the profile and inputs must be known at the land surface. The third method involves the total amount of tracer in a profile.

$$T(t) = \int_{z=0}^{z=\infty} T(z)\theta(z)dz$$

Where $T(t)$ is the total amount of tracer in the profile, $T(z)$ is the tracer concentration of the water in the unsaturated zone at some depth z , and $\theta(z)$ is the volumetric water content. This last method, also known as the tracer mass balance, is most often used with chloride.

The chloride mass balance method (Allison, 1978) has been used successfully in many different studies. This method is particularly useful, because accuracy is inversely dependent on the moisture flux. Unlike many of the physical methods in which the accuracy of the moisture flux calculations decreases as the flux decreases, the flux estimates from the chloride mass balance approach become easier to measure, as the net downward moisture flux decreases. Most plants do not take up significant amounts of chloride from the soil water so Cl^- is concentrated within the root zone by evapotranspiration. The evapotranspiration efficiency of desert plants is so complete that one study (Allison, 1983) documented a change in recharge from $<0.1\text{mm/yr}$ to over 3mm/yr following the clearing of native vegetation for agricultural purposes.

There are several assumptions used in the chloride mass balance method. The method assumes vertical piston flow and no internal sources of chloride. The vertical flow assumption appears to be valid on slopes less than 2%. For slopes greater than this, the chloride mass balance method underestimates moisture flux because lateral flow is omitted (Scanlon, 1991). Chloride input occurs at land surface as both dry-fall and in precipitation. It is assumed that any internal production of chloride can be distinguished from an atmospheric source by its distinctive Cl/Br ratio and or isotopic signature. The piston flow assumption is a difficult one to assess. It most likely depends on the scale, which is being considered. Near surface where desiccation cracks develop, piston flow is probably not a good assumption. However, in arid regions the matric potentials are generally very low so it is more likely that capillary forces will tend to diffuse

unstable behavior provided fluxes are small. A large uncertainty in the method is the ability to actually measure the chloride in pore water (Murphy, 1996). It is assumed that the chloride is so soluble that it will all come out into solution. This, however, ignores the fact that chloride can often be tied up in a caliche matrix that surrounds individual grains. The degree to which the chloride is tied up can vastly alter the results of a chloride mass balance study. The chloride mass balance method, and other tracer methods are only effective if the lithology and the dynamics of the flow system are carefully considered. If used properly they represent less uncertainty than physical methods of measuring recharge. The use of the CMB method hinges on an accurate estimate of chloride input into the system. The estimated ages and moisture flux are very sensitive to this parameter. There are several approaches for determining the chloride deposition flux. First, current chloride deposition can be measured using precipitation collectors and bulk samplers. This current rate then can be extrapolated over the range of the profile. This is probably the least accurate as it assumes no chloride deposition changes through time. Other methods for determining chloride flux involve the use of isotopes and are discussed below.

Paleorecharge and Paleoflux Estimates from Vadose Zone Profiles

The basic assumption for using tracers in arid vadose zone profiles is that velocities are slow enough that water at depth entered the system under markedly different climatic boundary conditions than today (Phillips, 1994). As a result the distribution of soil water tracers, such as isotopic tracers, may be very useful in reconstructing paleohydrologic conditions over time scales comparable to, if not longer than, lake sediment records. Initial studies have used only chloride as a tracer while later studies have used ^{36}Cl , ^{18}O , Deuterium, and other isotopes to further constrain their estimates of boundary conditions.

Recent studies by (Phillips, 1988; Phillips, 1994; Scanlon, 1991; Tyler et al., 1996) have shown the utility of using the distributions of chloride and ^{36}Cl in vadose zone profiles to infer paleorecharge under past hydrologic regimes. A key element to these analyses is the estimation of soil water ages. The CMB method can also be used to solve for the age of the soil water at a given depth, such that the age $A(z)$ at any depth z in the soil profile is taken as the cumulative mass/unit area of chloride (θC_{sw}) from the surface to depth z^* divided by the annual chloride deposition rate ($P(C_p + D)$ or PC_{eff}).

$$Age(z^*) = \frac{\int_{z=0}^{z=z^*} \theta C_{sw} dz}{PC_{eff}}$$

This equation presumes a constant rate of chloride deposition. The concentration of chloride in

precipitation is primarily a function of location with respect to major ocean bodies and is unlikely to have changed much over the period in question. So any changes in chloride deposition rates would be due to changes in precipitation rates or changes in the eolian chloride component during periods of lake desiccation and high winds. Neither of these is well quantified and the CMB method has been justly criticized for this uncertainty. Common techniques are either to use current flux rates to conservatively estimate chloride input or to use various extrapolation techniques to account for increases during past glacial periods. A possible alternative solution to this problem is natural variations of ^{36}Cl in the profile.

Before using this technique it is necessary to understand the mechanics of ^{36}Cl production. Chlorine-36, an isotope of chlorine, is primarily produced in the upper atmosphere by the bombardment of ^{40}Ar by cosmic radiation. The rate of production of ^{36}Cl varies with the intensity of the earth's magnetic field. This is illustrated in the figure 1 below (Plummer, 1996).

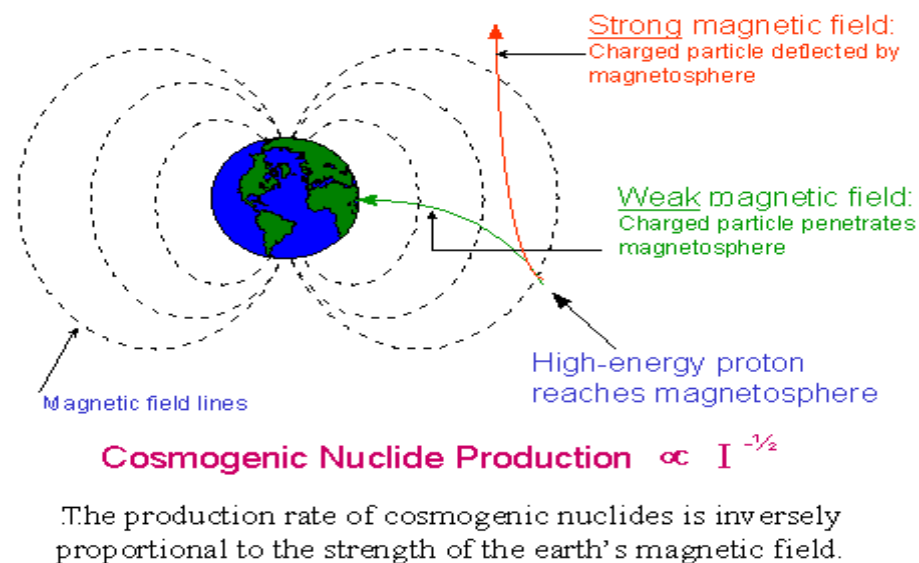


Figure 1

This variation in the production of ^{36}Cl can be traced through records such as those preserved in packrat middens and vadose zone profiles. For a historical average of chloride deposition it is possible to divide the natural ^{36}Cl fallout at the site, which is only a function of latitude, by the $^{36}\text{Cl}/\text{Cl}$ ratio in a sample from the site. Ratios that diverge from the expected ratio can be attributed to increased precipitation, and thus additional chloride, or increases in eolian deposition during periods of increased atmospheric dust. Chloride concentration in the precipitation is primarily a function of distance from the marine source. As this relationship has not changed dramatically during the Pleistocene, the assumption that chloride concentration in precipitation remains fairly constant is probably justified (Dettinger, 1989). A study in eastern Washington checked the $^{36}\text{Cl}/\text{Cl}$ -calibration method with a record that had essentially been reset during Lake Missoula floods 14,000 years ago. They found very good agreement with the Cl deposition rates determined by averaging this known record with those obtain from the $^{36}\text{Cl}/\text{Cl}$ ratios (Murphy, 1996). As mentioned above, one factor that could skew the chloride deposition rates is additional eolian input during interpluvial periods of desiccated lakes in the Great Basin.

This may be resolved with ^{36}Cl isotopes. Airborne lacustrine sediments have a significantly different $^{36}\text{Cl}/\text{Cl}$ signature than Cl of marine origin (Phillips, 1994).

Study Plan

The following study plan outlines how I will go about satisfying the various research objectives that were outlined earlier.

The first step in the paleoclimate reconstruction is to develop a new vadose zone chloride record for the Yucca flat core. The top 40m of UE-6e was never cored. We have gone back and drilled another hole adjacent to the original hole, designated UE-6s. This hole lies 60m to the south of UE-6e and penetrates 50m of alluvium. The 10m of overlap show similarities between the two cores. These two records combined should provide a complete record for use in paleoclimate reconstruction. Samples were collected from Ue-6e and newly drilled Ue-6s. The samples were analyzed for chloride, stable isotopes, water content and other soil parameters. They were also selectively analyzed for ^{36}Cl . The initial ^{36}Cl analysis will be used to pinpoint the best locations for future sampling. This reconstructed record will eventually be used to test the assumption of constant chloride deposition through time, the central tenet of the chloride mass balance method. The paleorecharge record as inferred from the chloride mass balance method will be compared to existing records.

In the summer of 1998 we conducted a supplemental drilling program on two transects adjacent to the UE-6s corehole. The purpose of these shallow cores was to look into the spatial heterogeneity of near surface properties, such as water content, total soil water potential and chloride content of the soil water. These cores were hand-augured down to a targeted depth of 5m. This will provide greater certainty in substituting the top portion of UE-6e with the newly drilled UE-6s. These shallow cores may also provide some additional information on preferential flow in the near surface.

As noted above, the chloride mass balance method critically relies on accurate estimation of the chloride flux to land surface. Part of this project will be to review all of the existing literature on Cl flux to better estimate this value and its possible ranges. Of particular interest are changes in chloride deposition during the late Pleistocene and Holocene transition. This was a period where both precipitation and eolian transport of dust changed dramatically based on ice core records (Biscaye, 1997; Yung, 1996). What can be learned from the literature about changes in chloride deposition during this period? To supplement the information in the literature, we have installed some chloride collectors based on a design developed by James Moore (1997). These collectors have been installed at a site in New Mexico and at a site in Yucca Flat adjacent to the corehole. Plans exist to install additional collectors at a site in the Sand Spring Mts. down wind from desiccated Lake Lahontan. These collectors will better constrain current chloride deposition rates and give us a backdrop from which to measure variations. They should also provide some information about eolian transport in three very different geographic settings.

When all of the ^{36}Cl sampling is complete, I plan to work with the group at New Mexico Tech to reconstruct a chloride deposition rate from that record. It has been shown in several studies that

the ^{36}Cl production rate, which is highly dependent on the strength of the magnetic field, can be reconstructed (Baumgartner, 1998). This record can be augmented within the working time scale of ^{14}C by comparing records that contain both ^{36}Cl and ^{14}C . Examples of such records are pack rat middens and groundwater (Plummer, 1997). The reconstructed record will be compared to existing local records. $^{36}\text{Cl}/\text{Cl}$ ratios that are out of phase with the expected ratio can be assumed to be periods of increased or decreased relative chloride deposition. This reconstructed record will help to better constrain the paleorecharge conclusions from the chloride mass balance method. Michelle Walvoord at New Mexico Tech has also been modeling the transport of chloride and ^{36}Cl in the unsaturated zone. I have been assisting in the calibration of this model. The results of the model can be used for this phase of the project to show the time scale and dynamics of presumed isolated nature of the tracer.

This project not only seeks to improve the utility of the CMB method but also to develop new ions for use as natural tracers in the vadose zone. The most likely candidate at this point is nitrate. It is produced, or fixed, often in large quantities, by plants and algae living on the surface or in the near surface environment. Once NO_3 moves past the root zone, it should be a reasonably conservative tracer. It also exists in concentrations that are often higher than those of chloride. The other plus is that this application is not dependent on atmospheric deposition but is only dependent on plant productivity. Changes in atmospheric nitrogen are small on this time scale. The use of nitrate as a tracer has been discussed as an option by W.M. Edmunds of the British Geological Survey (Edmunds, 1997). Nitrate has been found in high concentrations in the soils at the NTS and in other Nevada valleys. It is postulated that the primary production is from cyanobacteria in cryptogamic crusts. Although nitrate is not completely conservative, it is nearly so once it gets beneath the root zone into the largely inorganic and aerobic unsaturated zone. This type of environment is consistent with most of the profiles that encompass this study. The molecular diffusion coefficients of NO_3 and Cl^- are similar. With the low soil water flux expected at these sites, NO_3 may well prove an effective tracer. Nitrate profiles can be developed just as the chloride profiles were. The use of additional tracers will provide additional collaboration for the paleorecharge record that we seek.

Preliminary Data: Frenchman Flat

A similar type of analysis was done on the PW series wells that were drilled in the vicinity of the proposed site of the Radioactive Waste Management Site (RWMS) in 1992. (Tyler, 1996). These wells were drilled as subsurface characterization holes. Soil samples were taken and eventually analyzed for chloride, ^{36}Cl , stable isotopes and ^{14}C . A reconstruction of the recharge history over the last 120,000 was completed from those wells. There was considerable variation in the chloride profiles from the three different wells, despite the fact that they were all within 3 km of each other. This was attributed to differences in surface topography, which could result in different runoff regimes. However, this analysis was done without the benefit of a reconstructed chloride flux record. A constant chloride flux was assumed and a value slightly higher than the present level was used. Part of this proposal is to go back and look at that record again to determine if there are significant changes in the regional chloride flux history. The other part of the project is to apply the reconstructed flux rates over time to the new record from wells UE-6e and UE.6s. Many of the same techniques that were applied to the PW series wells will be applied to this record. This will allow for comparison of the two records. Because the boundary

conditions at the two sites are thought to be similar, this comparison will also provide insight into the differences in transport processes resulting from different lithologies.

Site Description

Yucca Flat is a 1400km², alluvium filled basin located in the northeast corner of the Nevada test site, 150km north of Las Vegas, NV. Yucca Flat is bounded by the Ryolite Hills to the north, Ranier Mesa and Syncline Ridge to the west, the Mine and Massachusetts Mts. to the south and the Halfpint Range to the east. Alluvium filled basins at the test site represent ideal outdoor laboratories for vadose zone studies. The water table is often deeper than 250m. The region is arid with an average annual rainfall of about 150mm. The depth to bedrock often exceeds 600m. Because of the use of the test site for a nuclear testing program for the last 50 years, many cores have been drilled and the subsurface has been extensively characterized. One such core, designated UE-6e, was drilled in 1973 using a unique and experimental drilling method that should have preserved environmental tracers in the core. This method is ideal for our analyses because it used air and air foams, as the only drilling fluids and thus contamination of the core with water is minimal. The core has been archived in the USGS core library since that time in a manner that should be free of contamination. Another supplemental hole, designated UE-6s, was drilled as part of this project, in spring 1998. Both holes were drilled in the southern portions of Yucca Flat near the Yucca Lake/ playa area.

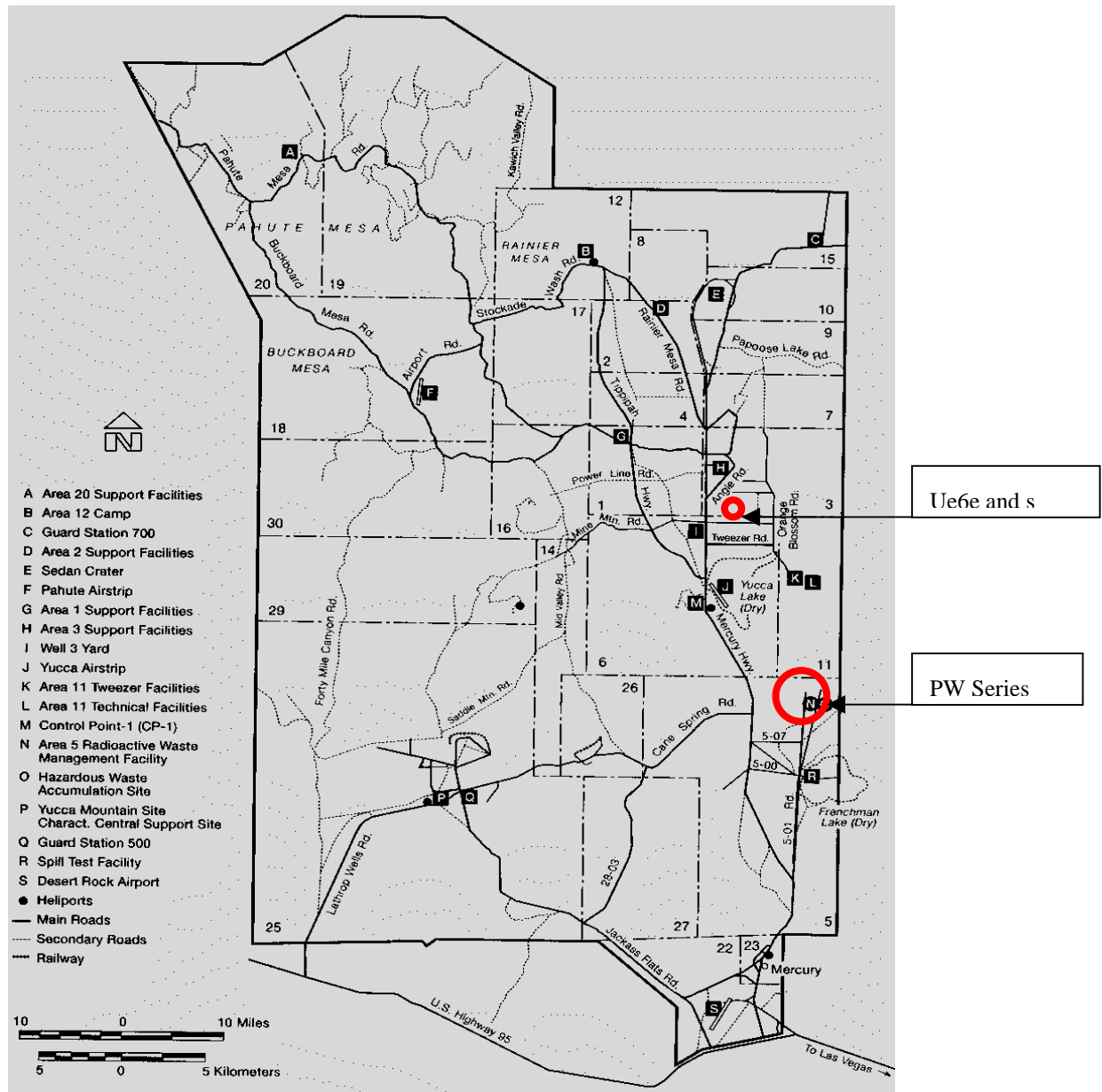
UE-6e- Geology and drilling history

Well UE-6e was drilled as an exploratory hole in Yucca Flat (N 248,107m, E 209,763m) at a surface elevation of 1,199.5m. The well was originally drilled as part of the subsurface characterization project for the Underground Testing Program and to explore the hydrogeology of southern Yucca Flat. The well intersected alluvium, volcanic tuff, and Paleozoic carbonate rocks. Our analysis will be limited to the alluvium section (417m) and several tephra lenses. The top 45m were never cored necessitating the drilling of UE-6s. In 1990, DRI and the USGS revisited UE-6e. The water table was measured at 460m and an obstruction was encountered at 704 m. The well was again visited in 1992 to attempt to clear the obstruction. This attempt failed and led to further collapse. The well is presently used only for water table monitoring.

UE-6s- Geology and drilling history

Much of the necessary tracer information for our project is located in the upper 40m. This portion of UE-6e was never cored. Our idea was to drill another hole 60m to the south of UE-6e (N 248,047m, E 209,763m). The distance of 60m was chosen to be close enough to be representative of the lithology and moisture flux history of UE-6e, but far enough away to assure

we were off the drilling pad and disturbed area from the original UE-6e excavation. The original drilling specifications for UE-6e called for a drilling pad extending 40m in all directions. 60m should put the hole safely off the drilling pad and any other drilling disturbances such as vehicles and drill pad irrigation. A depth of 50 m was chosen to allow some overlap with UE-6e for comparison of lithologies and chloride contents. The hole was drilled in February 1998 using the hollow stem auger method and near continuous coring.



Map of Nevada test site showing location of PW wells, UE-6e and newly drilled UE-6s

Project Schedule/ Progress

Drilling of well UE-6s and sampling of the cores has already been completed. The ^{36}Cl analyses are underway and additional sampling will be done, as the results require. Last summer chloride collectors were installed at the NTS to supplement those already installed in New Mexico. The installation of collectors at a site in the Sand Spring Mountains down wind from the desiccated Lahontan playa is slated for this spring. Last summer collection of data for a spatial variability study in the region of wells UE-6e and 6s was started to test our assumption of well UE-6s being representative of the top portion of 6e. This spring will see the compilation of these data and the reconstruction of the paleoclimate and paleorecharge records in the Yucca Flat area. These reconstructed records will be compared to the PW series records and other regional records and eventually become part of the larger regional paleoclimate reconstruction. The immediate goal of this project is to produce a paper suitable for publication in a scientific journal. The eventual goal of this project is to contribute to the global paleoclimate record and to be of use for decisions regarding the siting of waste facilities and water supply issues in the arid west.

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