Study Plan for
Study 8.3.1.2.2.2

Water Movement Tests

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Revision 0

January 1989

U.S. Department of Energy
Office of Civilian Radioactive Waste Management
Washington, DC 20585

Prepared by
Los Alamos National Laboratory

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YUCCA MOUNTAIN PROJECT

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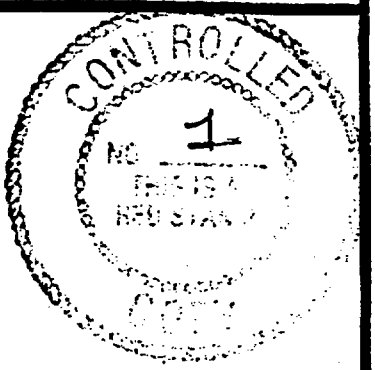

Study Plan Number 8.3.1.2.2.2

Study Plan Title Water Movement Tests

Revision Number R0, January 1989

Prepared by:
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January 1989

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WATER MOVEMENT TEST

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ABSTRACT

The water movement tracer test is designed to produce information derived from isotopic measurements of soil and tuff samples collected from Yucca Mountain that is pertinent for assessing the performance of a nuclear waste repository. Measurements of chlorine isotopic distributions will help characterize the percolation of precipitation into the unsaturated zone. The chlorine-36 in the unsaturated zone occurs from atmospheric fallout of chlorine-36 produced by cosmic-ray secondaries reacting with argon-40, and, to a lesser extent, with argon-36 and as global fallout from high-yield nuclear weapons tests conducted at the Pacific Proving Grounds between 1952 and 1963. When chloride ions at the surface are washed underground by precipitation, the radioactive decay of the chlorine-36 in the chloride can be used to time the rate of water movement. The chlorine-36 half-life of 301,000 yr permits the detection of water movement in the range of approximately 50,000 to 2 million years. These data are part of the input for developing numerical models of groundwater flow at this site.

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STUDY PLAN FOR WATER MOVEMENT TEST
Site Characterization Plan Study 8.3.1.2.2.2

1.0 PURPOSE AND OBJECTIVES OF STUDY

1.1 Purpose

This study consists of a single activity (8.3.1.2.2.2.1). The purpose of this study is to determine the rate of water movement downward through the unsaturated zone beneath Yucca Mountain, using measurements of chloride concentrations and chlorine isotopic compositions in samples of soil and tuff collected as part of the site characterization program. The objective of one phase of this study is to determine an upper bound on the amount of water that enters Yucca Mountain by infiltration. The objective of the second phase is to time water movements in the unsaturated zone by measuring chlorine-36/total chlorine ratios in samples that will be collected as the exploratory shaft is mined. The 301,000-yr half-life of chlorine-36 is useful for tracing water movements between 50,000 and 2,000,000 yr and is most useful between 100,000 and 1,000,000 yr.

The data from this test will be used as part of the information required by the Yucca Mountain Project (YMP) to calculate releases to the accessible environment. These data will help establish an accurate model of the hydrologic characteristics of the unsaturated zone at Yucca Mountain. The hydrologic model will be used to compute radioactivity releases to the accessible environment as part of the repository performance assessment.

Another use of the data will be to estimate the rate of technetium-99 movement through the unsaturated zone. The technetium-99 is likely to be in the chemical form TcO_4^-, which, like chloride, is a nonsorbing geochemical form. The rate of movement of technetium will be no faster then the chlorine-36 rate of movement.

1.2 Resolution of Performance Issues

The rationale for the YMP site characterization program is presented in Section 8.1 of the YMP Site Characterization Plant (SCP) (DOE, 1988). The issues-based strategy was guided first by an issue identification procedure and then by performance allocation to define the activities needed to resolve the issues. The issues were divided into performance issues and design issues. The work in this study plan applies only to performance issues.

The primary issues that will use the data from this study are the following:

Issue 1.6: Will the site meet the performance objective for pre-waste-emplacement ground-water travel time as required by 10 CFR 60.113?

Issue 1.1: Will the mined geologic disposal system meet the system performance objective for limiting radionuclide material to the accessible environment as required by 10 CFR 60.112 and 40 CFR 191.13?

The measurements described in this study plan do not directly contribute to any of the performance parameters (discussed in SCP Sections 8.3.5.12 and 8.3.5.13) that will be used to calculate ground-water travel time and total system performance. Those parameters are generally hydrologic properties of the rocks and fluids that will be used to construct models of flow and transport at Yucca Mountain. Instead, as noted in SCP

Section 8.3.5.12 (Issue 1.6), the isotopic measurements described here will provide important information that may confirm or invalidate model predictions of ground-water velocities and fluxes in the unsaturated zone. Section 2.2 of this study plan discusses the interpretation of the isotopic data with respect to the determination of rates of water movement.

1.3 Regulatory Rationale and Justification

As indicated in the discussion of performance issues, this study could provide support concerning compliance with several key regulations. The Department of Energy's siting guidelines [10 CFR 960.4-1-2(b)(1)] specify that a pre-waste-emplacement ground-water travel time greater than 10,000 yr along any path of likely radionuclide travel from the disturbed zone to the accessible environment would be a favorable condition. The long travel times that this test can measure will bear directly on the evaluation of this favorable condition. The data may be used to determine the nature and rates of hydrologic processes that have occurred during the Quaternary Period, which are specified in 10 CFR 960.4-2-1(b)(2). The sampling procedure for this test (see Section 3.1 below) is designed to detect stratigraphically influenced changes in the rate of water movement through the unsaturated zone. Such changes, if detected, will be used for validating the modeling of the geohydrologic system that is required in 10 CFR 960.4-2-1(b)(3). All of the above information will be used to support the higher level findings on the geohydrology disqualifying and qualifying conditions specified in 10 CFR 960.4-2-1(a) and (d).

The second major source of regulatory requirements is the technical criteria of the US Nuclear Regulatory Commission (NRC). In 10 CFR 60.113(a)(2), the NRC requires that the pre-waste-emplacement ground-water travel time along the fastest path of likely radionuclide travel from the disturbed zone to the accessible environment shall be at least 1,000 yr. The NRC also specifies that the geologic repository system performance following permanent closure shall conform to the applicable environmental standards for radioactivity as established by the Environmental Protection Agency (10 CFR 60.112). The hydrologic flux in the unsaturated zone is an important component of the total system performance assessment, and the measurements in this study are part of the site characterization process to determine the unsaturated-zone flux.

2.0 RATIONALE FOR USE OF CHLORINE-36 AND CHLORIDE TO TRACE WATER MOVEMENT IN THE UNSATURATED ZONE AT YUCCA MOUNTAIN

2.1 Estimates of Rates of Water Movement from Hydrologic Data

Determining the rate of water movement through the unsaturated zone at Yucca Mountain is one of the most important tasks for assessing the future performance of a nuclear waste repository, but it is a very difficult task. Montazer and Wilson (1984) discuss the data on water movement available through 1984. Their hydrologic terminology is used in this study plan, as it is in the YMP SCP (DOE, 1988). Estimates of downward flux through the potential repository host rock, the moderately to densely welded portion of the Topopah Spring Member of the Paintbrush Tuff formation, range from 1×10^{-7} to 0.2 mm/yr. These estimates were based on hydraulic gradient and effective permeability data derived from one borehole and from cores recovered from holes at more than one location. Analyses of geothermal heat-flux data from the same tuff unit indicate that the net hydrologic flux may be upward at a rate of 1 to 2 mm/yr, possibly from vapor-phase transport. Montazer and Wilson (1984) state that the hydrologic flux through the tuffaceous beds of Calico Hills, which underlie the host rock, is likely to be variable, but less than about 0.006 mm/yr downward, as estimated from

measurements of effective hydraulic conductivities from core samples that included the zeolitic facies of this unit.

The hydrologic flux estimates outlined in the preceding discussion do not give a definitive picture of the rate of water movement to be expected through the unsaturated zone. If the flux through the Topopah Spring Member is assumed to be about 0.2 mm/yr downward, then estimates of ground-water travel time through the unsaturated zone are greater than 100,000 yr. Such estimates indicate that chlorine-36 techniques would provide data useful for hydrologic modeling.

2.2 Radiometric Determination of Rates of Water Movement

Direct measurements of the rate of water movement through the unsaturated zone would be of great value. Radiometric methods are one potential technique for making such measurements. Determinations of tritium and carbon-14 in the unsaturated zone are discussed in the hydrochemistry study plan (8.3.1.2.2.7). The 5,730-yr half-life of carbon-14 permits dating to ages of 60,000 yr under favorable conditions. Other radiometric dating techniques have been considered. A survey of such techniques given by Phillips (1984) indicates that radiometric dating with iodine-129 may be possible for waters older than 10^6 years. The applicability of this technique to waters moving through the unsaturated zone at Yucca Mountain may be limited by the lack of a useful iodine-129 source. The technique chosen to determine the water travel time through the unsaturated zone in this study is the measurement of chlorine-36 as a function of depth. Chlorine is deposited globally both in precipitation and in dry fallout. The source of most of the chlorine in this fallout is sea salt lofted into the troposphere by surface winds. A very small fraction of chlorine atoms in the fallout consists of chlorine-36, which results from cosmic ray reactions with argon in the atmosphere. The chlorine-36 half-life of 301,000 yr is appropriate for the travel times calculated from the hydrologic data. Geochemical properties of chlorine make it a useful tracer of subterranean water movements (Bentley et al., 1986). Chlorine is so electronegative that it exists as the nonvolatile chloride ion under most geohydrologic conditions. Bentley et al. (1986) state that chloride ions are among the least sorbed ions on solid surfaces because of their negative charge and small radius. Measurements of chlorine-36 discussed in Bentley et al. (1986) indicate the validity of using this technique for tracing water movements over long times.

At Yucca Mountain the technique discussed in this study plan will measure chloride carried into the tuff by meteoric water and now present in pore water or on the surfaces of the tuffs. This chloride traces the movement of water in the liquid phase through the unsaturated zone. Chlorine originally present in the tuffs (the chlorine-36 component of which should have decayed to negligible amounts at the present) may be included in the samples. Most of this chlorine may be chemically bound. Short leaching times should help minimize the introduction of this chlorine into the samples for the chlorine-36 measurements.

Measurements to aid in interpreting the chlorine-36 data include chloride concentration with depth and stable chlorine isotope ratios. The chloride concentration profile is expected to be relatively uniform, if the water movement in the unsaturated zone is uniform. Deviations in the chloride concentration profile may result from nonuniform evaporation or from mingling of waters caused by localized cross-cutting features. These data may help confirm patterns of nonuniform flow in the chlorine-36 data. The stable chlorine isotope ratio measurements may indicate that detectable differences can be observed in the chlorine-35/chlorine-37 ratio, depending on the origin of the chlorine. Such differences have been reported recently (Desaulniers et al. 1986). At Yucca

Mountain, the chlorine-35/chlorine-37 studies may show that chlorine of meteoric origin has one ratio whereas chlorine originally present in the tuff has a different ratio. The chlorine-36 content associated with chlorine of meteoric origin is the only quantity of interest. Measurements of chlorine-35/chlorine-37 ratios in the samples for chlorine-36 analysis can be compared with the ratio determined for chlorine of meteoric origin and with the ratio measured for chlorine from tuff to correct the chlorine-36/chlorine ratio to that which should be observed in chlorine of meteoric origin.

The specific activity of chlorine-36 in the environment is so low that measurements of the chlorine-36 isotopic abundance have not been feasible, with a few exceptions, until the recent development of tandem accelerator mass spectrometric analyses. The sensitivity of this technique currently is approximately one atom of chlorine-36 in 10^{15} atoms of chlorine. The sample size required for these measurements is 10 mg of chloride or more. The background chlorine-36/chlorine ratio in surficial deposits of alluvium at Yucca Mountain has been measured in this work to be approximately 5×10^{-13}, which is more than 100 times greater than the limit of this technique. Any samples collected in which chlorine-36 has undergone detectable radioactive decay will result in chlorine-36/chlorine ratios lower than that of the cosmogenic background. Radiometric determinations are limited to samples that have been isolated from isotopic exchange with cosmogenic sources of chlorine-36.

Several recent studies suggest that chlorine-36 may be a suitable indicator of the rate of water movement. Andrews et al. (1986) used the chlorine-36/chlorine ratio to estimate ground-water residence times at the Stripa mine in Sweden and formulated conceptual ground-water flow paths to explain the evolution of the ratio.

Norris et al. (1987) used measurements of chlorine-36 in soil samples from two locations near Yucca Mountain to determine the infiltration of precipitation during the past quarter century and to examine the differences in surficial hydrologic infiltration between the two sites. The source of the chlorine-36 measured in this work was not from cosmic ray reactions with argon in the atmosphere. Instead, the chlorine-36 was deposited globally as fallout from high-yield nuclear weapons tests that were conducted at the surface of the Pacific Ocean between 1952 and 1962. The data can be interpreted in terms of an infiltration rate at Yucca Wash, located to the east of Yucca Mountain, of 1.8 mm/yr during the past quarter century. This value represents an in situ measurement of flux that is valuable in establishing the upper bound for the amount of water flowing downward through the unsaturated zone.

The mechanisms of water transport in the unsaturated zone are of interest for appropriate modeling of radionuclide transport. However, the chlorine-36/chlorine data from this study are unlikely to discriminate between fracture flow and porous flow because the chloride that is measured can result from water flow by a combination of mechanisms. The chlorine-36/chlorine data may shed light on the mechanism of water flow only if rapid fracture flow dominates to the extent that bomb pulse chlorine-36 is observed at great depths or if the hydrologic flow is so slow that hydrologists would describe the mechanism as porous flow.

2.3 Constraints on Chlorine-36 Studies

Analytical and Sampling Constraints

The specific activity of chlorine-36 present in tuff samples from Yucca Mountain, as in typical samples from other environments, is too small to permit direct measurements of the radioactive decay of chlorine-36. Conventional mass spectrometry can be used to

measure chlorine-36/chlorine ratios as low as 10^{-10}, but the ratios in Yucca Mountain are approximately 5×10^{-13} for samples with contemporary cosmogenic chlorine-36. Therefore, the only feasible technique for this study is tandem accelerator mass spectrometry. The 10 mg of chloride required for the chlorine-36/chlorine measurement will be leached from crushed tuff samples, as described in Section 3 of this study plan. The chloride content of tuff samples from the Paintbrush Tuff formation at Yucca Mountain necessitates collecting about 40 kg of rock per sample. The collection process should involve little or no contact of water with the sample to be collected, to avoid inadvertent removal of the chloride before the leaching process that is part of the analysis procedure.

These constraints led to the necessity for using tuff samples collected as the exploratory shaft is mined. Some water will be used in the drilling and blasting operations, but large rubble pieces can be chosen to minimize any wetting of the major part of the rubble volume. This study could not be performed with core samples from conventional surface-based drilling, in which water is used as the lubricant for the drilling bit.

Samples will be collected from the exploratory shaft as shown in Figure 1. The sampling locations indicated were selected on the basis of stratigraphic data from USW G-4. The strata might be factors in controlling the downward movement of water through the unsaturated zone penetrated by the exploratory shaft. The chlorine-36 half-life constrains the interpretation of the data to water movements that have taken longer than 30,000 yr, because too little chlorine-36 decay has occurred to be detectable for younger times. Measurements of samples collected from the upper part of the exploratory shaft may give no useful data because of this threshold limitation. The half-life of chlorine-36 also constrains the measurements of age differences between samples to those that are greater than 30,000 yr. The data from deeper samples should provide information about water movement in the tuff above the repository horizon as well as below it.

The impact of this study on the potential repository site will be negligible. Each sample will require that about 40 kg of rubble be used for analysis. Preliminary plans call for the water used to construct the exploratory shaft to be tagged with 20 ppm of bromide. This will permit detection and correction for approximately 10 ppm of chlorine in that water. Some samples for chloride profile studies may be at shallow depths (<10 m) in locations of potential infiltration, but these locations will be selected after a field study of Yucca Mountain.

The analytical techniques for this study are available and are described in Section 3 of this plan. Cosmogenic chlorine-36 in samples of Yucca Mountain alluvium has been measured (Norris et al. 1987). The tandem accelerator mass spectrometric measurements have to be performed when the apparatus is available. There are a sufficient number of tandem accelerator users that a wait of several months can be expected. Exploratory shaft operations are not affected by this wait. The time required for interpretation of the chlorine isotope data does not constrain any other activity.

Chlorine-36 can be produced underground from neutron capture by chlorine-35. The neutrons result from the presence of uranium and thorium and their decay chains. Some neutrons come from spontaneous fission of the first members of the decay chains. The remainder are produced by (α,n) reactions on light elements. Calculations will be performed to determine whether in situ production of chlorine-36 will constrain the interpretation of the data from this test.

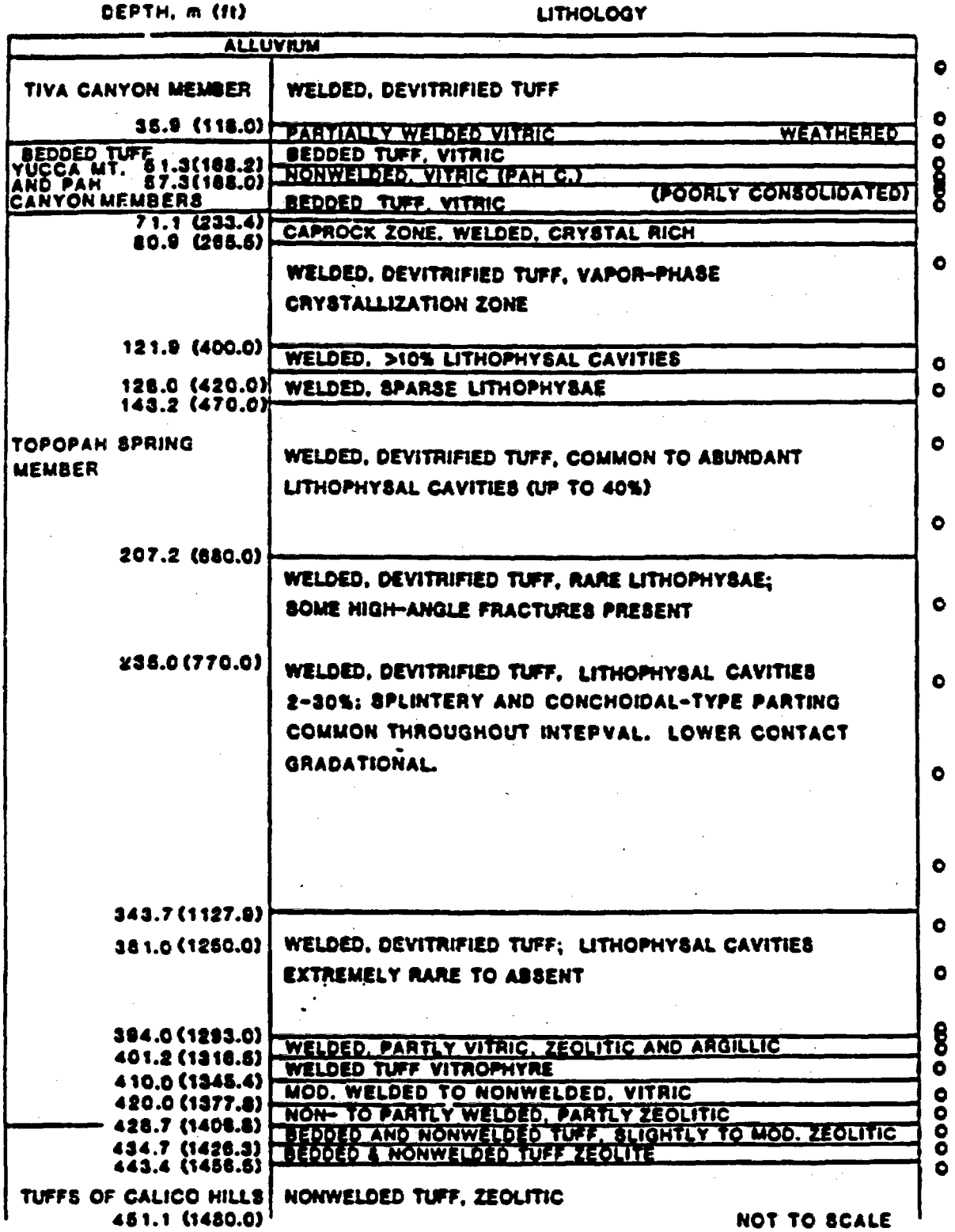

Figure 1. USW G-4 stratigraphic column with proposed chlorine-36 sampling locations shown as circles. Total sampling depth will be constrained by the depth of the Exploratory Shaft Facility.

2.4 Potential for Interference with the Chlorine-36 Studies

The interferences to which this study is susceptible from exploratory shaft operations are due to the use of water, which has been discussed above, and the introduction of chlorine that could alter the in situ chlorine-36/chlorine ratio. The explosives used in mining the exploratory shaft may contain chlorine with negligible chlorine-36. Samples for this study will be collected to minimize the possibility of contamination from chlorine in the explosives.

3.0 DESCRIPTION OF CHLORINE-36 STUDY

The key parameter to be measured in this study is the ratio of chlorine-36 to total chlorine as a function of depth in the exploratory shaft at Yucca Mountain. Location of the exploratory shaft is shown in Figure 2.

3.1 Sample Collection

Most of the sample collection will be performed as the exploratory shaft is mined. The principal borehole (USW G-4) stratigraphy was used to select sampling depths on the basis of lithology and stratification that might be significant for hydrologic characteristics. When there were no stratigraphic features to guide the selection of sampling intervals, the maximum interval between samples was set arbitrarily at 30 m (100 ft). The locations of samples are shown in Figure 1. The depths of sampling are subject to minor modifications to account for the dip of the beds between USW G-4 and exploratory shaft 1 (ES-1) and because the stratigraphy at ES-1 may not be exactly the same as that at USW G-4. The data from the Shaft-Wall Mapping Test (see geologic study plans) will be monitored to ensure that the blasting rounds will be sampled where significant lithologic changes occur.

Sampling will be performed in accordance with the procedures that will be developed by the YMP Sample Overview Committee.

After each designated round, a geologist will descend to the working face of ES-1 to select rubble pieces larger than 130 mm (6 in.) in diameter for transport to the surface before the customary washdown. At the surface, the rubble may be segregated into a special container. A 208-L (55-gal) barrel will be packed with 100 to 200 kg (220 to 440 lb) of rubble and labeled with the depth from which the rubble was collected.

Accidental contamination of samples with chlorine-36 is not expected to be a problem. Chlorine-36, unlike carbon-14, is not transported in the vapor phase. No special atmospheric protection is necessary during sample preparation. Quantities of chlorine-36 produced from nuclear reactors or particle accelerators are not stocked routinely where the chlorine-36 sample preparation is likely to be performed.

Most chlorine that might accidentally contaminate the samples from ES-1 would come from chloride ions in the Well J-13 water used in construction and from chlorine in the explosives that will be used during shaft-sinking operations. Therefore, the selection of larger pieces of rubble and the postponement of the customary washdown after blasting are two steps taken to mitigate potential sample contamination problems. The chloride content of Well J-13 water is approximately 10 mg/L. The water used underground will be tagged with a sodium bromide tracer. The bromide concentration of the Well J-13 water will be increased to 20 ppm, which is $\geq 10^3$ times the natural bromide concentration. The bromide content of the rubble selected for chlorine-36 analysis will

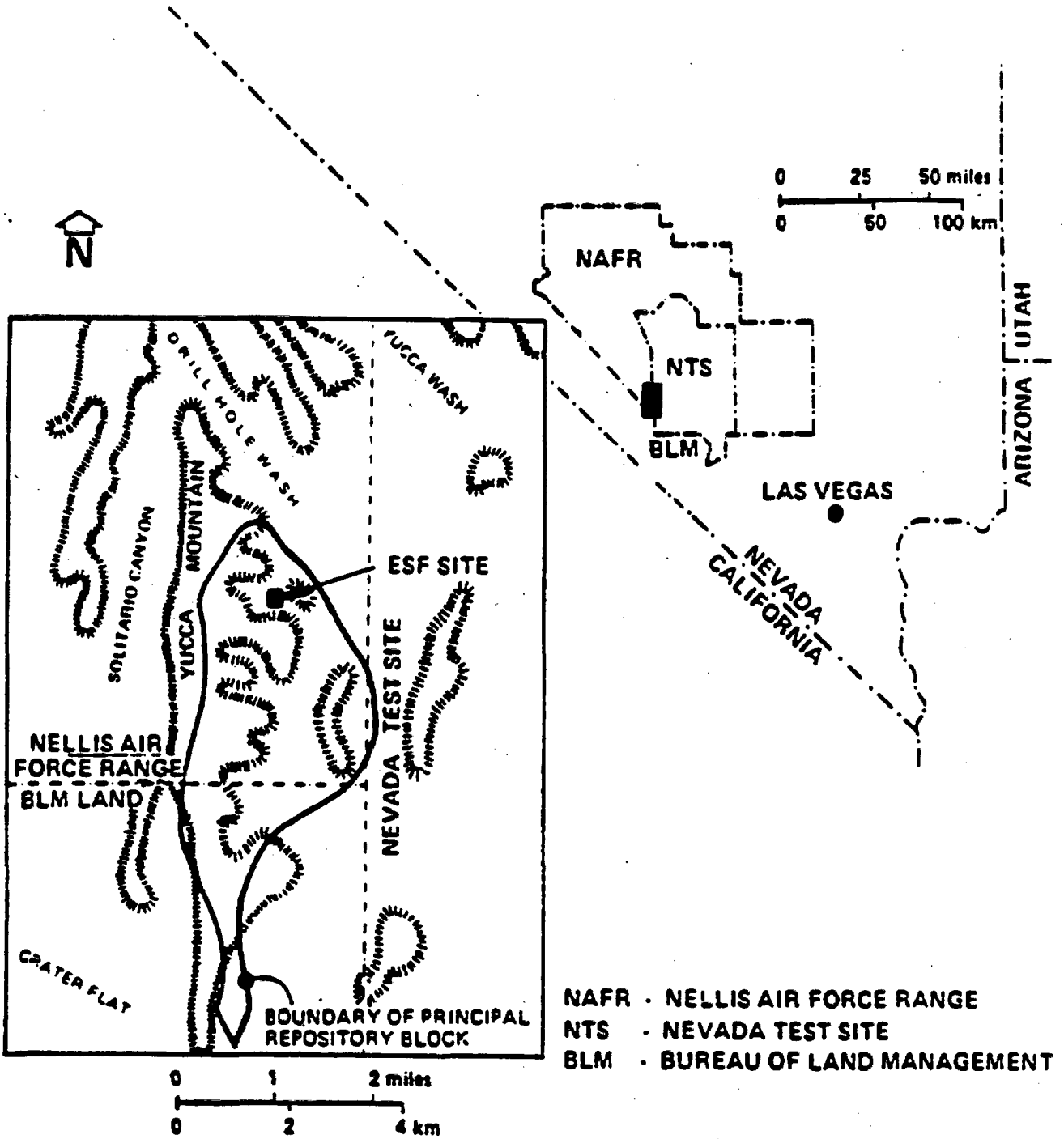

Figure 2. Relationship of the exploratory shaft facility to the Yucca Mountain site.

be measured to permit the calculation of the chloride that might have been introduced from Well J-13 water. Finally, a chlorine analysis of all the explosives used during the mining operations will be obtained and used to set a bound on the maximum amount of chlorine that could contaminate the samples from explosives.

The introduction of chlorine from Well J-13 water and from explosives is expected to result in at most a minor perturbation in the data because the quantities of chlorine are small. Explosives customarily contain chlorine as a minor or a trace constituent. A problem more difficult to evaluate is the downward movement of water used in construction, which may leach chloride from tuff below the working depth before samples are collected for this test. Selecting rubble pieces with large diameters may ameliorate this problem, because water flow into the interior of an intact piece of tuff is slow. If the data indicate that leaching before sample collection is a problem, additional samples can be obtained by using horizontal dry coring techniques. The shaft liner and the surrounding tuff can be penetrated to a depth where the downward flow of water used in construction is unlikely to be a problem.

Some samples for measuring the chloride profile at depths to a few meters below the surface may be collected independently of the exploratory shaft mining operations, particularly if the hole for the exploratory shaft collar is excavated in a way that precludes obtaining samples at known depths. These data will aid in interpreting the chlorine-36 data by providing a source term for the expected quantity of meteoric chloride.

Any water encountered in sufficient volume during the construction of the exploratory shaft will be sampled and analyzed for chlorine-36. Pore water samples will be analyzed for chlorine-36 if sufficient volume can be obtained (see SCP Section 8.3.1.2.2.8, Hydrochemistry).

The Yucca Mountain Project is considering drilling deep boreholes with dry drilling techniques. Samples from various depths in these boreholes will be requested for chlorine-36 analyses. If the data from the first few boreholes indicate no significant variation with depth or with location, the sampling will be discontinued.

3.2 Measurement of Chlorine-36/Chlorine Ratio

The chlorine-36/chlorine ratio will be measured in the tuff samples collected from ES-1. A subcontractor will be chosen to prepare the rock samples and perform the analyses of chlorine-36, because a highly specialized technique is used (Elmore et al. 1984a). The rubble selected for the chlorine-36/chlorine ratio measurements will be crushed. The bromide content will be measured to determine the quantity of chloride from well J-13 water in the sample, and the crushed rock will be contacted with chloride-free water. Silver nitrate will be added to the water to precipitate about 50 mg of silver chloride. This precipitate will then be analyzed for the chlorine-36/chlorine ratio in a tandem accelerator mass spectrometer.

Descriptions of the apparatus and the techniques currently being used for tandem accelerator mass spectrometric analyses of chlorine-36 are given in Elmore et al. (1984a). Chlorine in the form of silver chloride is accelerated first as negative ions to eliminate interferences from argon-36, which does not form negative ions. The chlorine ions pass through an argon gas stripper in the center of the tandem accelerator. Ions with a charge of +7 are selected for mass analysis in a 90 degree magnetic analyzer. Chlorine-35 and chlorine-37 are measured in a multiplate gas ionization detector. Measurement of energy loss in this detector permits separation of chlorine-36 ions from

interfering sulfur-36 ions. Measurements of chlorine-36 with precisions as good as ±5% have been obtained. The data in this study plan, from analyses performed on a routine basis, are expected to be measured with a one standard deviation (σ) precision of ±10%.

3.3 Analysis and Interpretation of Isotope Data

Table 1 summarizes the parameters that will be measured in this study and provides expected values. The range of expected values for the chlorine-36/chlorine ratio is determined at the upper end by the contemporary quantity of cosmogenic chlorine-36 and at the lower end by the sensitivity of the tandem accelerator mass spectrometric technique. The bromide will be added as a tracer to tag the water used in the exploratory shaft; the maximum quantity added will be 20 ppm. The ion chromatography sensitivity limit is 20 ppb. The limit on the sample collection depth is determined by the total depth of the exploratory shaft. The final part of this test is the analysis and interpretation of the data.

TABLE 1

PARAMETERS MEASURED IN THIS TEST

Procedure	Parameter Measured	Expected Value
Tandem accelerator mass spectrometry	Chlorine-36/chlorine	1 to 600 x 10^{-15}
Ion chromatography	Total bromide	≤20 ppm
Linear measurement	Depth of samples in ES	≤457 m (<1500 ft)
ASTM mercuric nitrate titration	Chloride concentration	1 to 1000 mg/kg rock
Conventional mass spectrometry	Chlorine-37/chlorine-35	-1 to 3 per mil relative to standard mean ocean chloride

The techniques used to obtain the chlorine-36/chlorine isotope ratios from tandem accelerator mass spectrometry measurements are documented in a paper by Elmore et al. (1984b). That paper discusses the calculation of the ratio of chlorine-36 to chlorine-35 + chlorine-37 in the sample, in a National Institute of Standards and Technology (NIST) chlorine-36 standard, and in a reagent blank. The isotope ratios are corrected for mass fractionation, for background, and for interferences arising from the presence of sulfur-36. The NIST chlorine-36/chlorine ratios are used to normalize the ratios in the samples for inaccuracies introduced by the tandem accelerator. The value of the final chlorine-36/chlorine ratio in the sample is calculated as the mean of the corrected and normalized ratios from a sequence of measurements, weighted by the uncertainty of each determination.

Interpretation of the chlorine-36 data in terms of the rate of water movement through the unsaturated zone will require the consideration of processes that differentiate chloride movement from water movement. One process is that of anion exclusion. Positively charged components in the subterranean mineralogic environment can exclude

negatively charged ions, which causes the anionic tracer to move slightly faster than tritiated water (Daniels 1983). The second aspect of chloride movement that is important for hydrologic modeling is the nonvolatile character of these anions. If water movement through the unsaturated zone should be in downward pulses through the matrix in the liquid phase, followed by upward movement in the vapor phase, the chloride ions would move only in the liquid phase. The chlorine-36 decay data, then, might show an age that results from mixing chloride ions from more than one pulse. The data in this case still would be useful for calculating the average travel time of a radioactive waste nuclide such as technetium-99, which is expected to be transported in water as the TcO_4^- ion. Like chloride, the pertechnetate ion is both nonvolatile and nonsorbing.

A premise of data interpretation for this study is that the chlorine-36 fallout has been constant throughout the Quaternary Period. Experimental evidence bearing on this premise is being sought in ice cores from Camp Century, Greenland (Elmore et al. 1984a). A perturbation in the constancy of the chlorine-36 fallout occurred between 1952 and 1962. High-yield nuclear weapons tests at sea level in the Pacific Ocean resulted in significantly increased global fallout of chlorine-36. This "bomb pulse" has been used to measure infiltration that occurred during the past 30 yr into the top few meters of sandy loam in New Mexico (Bentley et al. 1986) and into alluvium at Yucca Mountain (Norris et al. 1987). Hydrologic flow at Yucca Mountain may occur through fractures in tuff and by some lateral flow, particularly through the Tiva Canyon and Pah Canyon Members. If chlorine-36 values higher than background are encountered in samples from the exploratory shaft, the data will be examined to determine if fracture flow or lateral flow might account for the inclusion of "bomb pulse" chlorine-36.

The ratio of chlorine-36 to chlorine as a function of depth will be correlated with the measured chloride concentration profile and with detailed data on fracture orientation and frequency. Regions of nonuniformity in the chloride concentration profile indicate nonuniform flow rates, and such nonuniform flow rates might be observable in the chlorine-36/chlorine profile. The data concerning fracture characteristics and distributions, to be provided by the US Geological Survey (USGS), will be studied as one possible explanation for the nonuniform flow.

3.4 Accuracy and Precision of Water Velocity Determinations

The chlorine-36 concentration data, when plotted against sample depth, provide a measure of water velocity down through the unsaturated zone, if a vertical flow path is assumed. The accuracy of the measurement from this test depends on the rate of water movement down through the unsaturated tuff. For velocities less than 5 mm/yr, the accuracy is high. It decreases for higher water velocities until the practical limit of the test is reached at water velocities of about 10 mm/yr. For water velocities greater than 10 mm/yr, the shorter lived nuclides discussed in Section 2.2 will be used. The following statistical analysis shows the sensitivity of this technique.

Power curves were used to estimate the probability of detecting small water movement velocities from chlorine-36/chlorine ratio measurements at the depths indicated in Figure 1. The analysis is based on a constant water flow rate throughout the volume penetrated by ES-1. A null hypothesis was tested, namely that the rate of water flow through the unsaturated zone is greater than or equal to 15 mm/yr (which corresponds to a slope of -1.5×10^{-4} m^{-1} or greater when the natural logarithm of the chlorine-36/chlorine ratio is plotted versus depth). Figure 3 shows a plot of velocity versus power. Power is the probability of rejecting the null hypothesis, when the null hypothesis is false. The significance level of 0.05 implies a 5% chance of erroneously rejecting the null hypothesis. The three curves in the figure are labeled with the $\pm 1\sigma$ values of the individual data points.

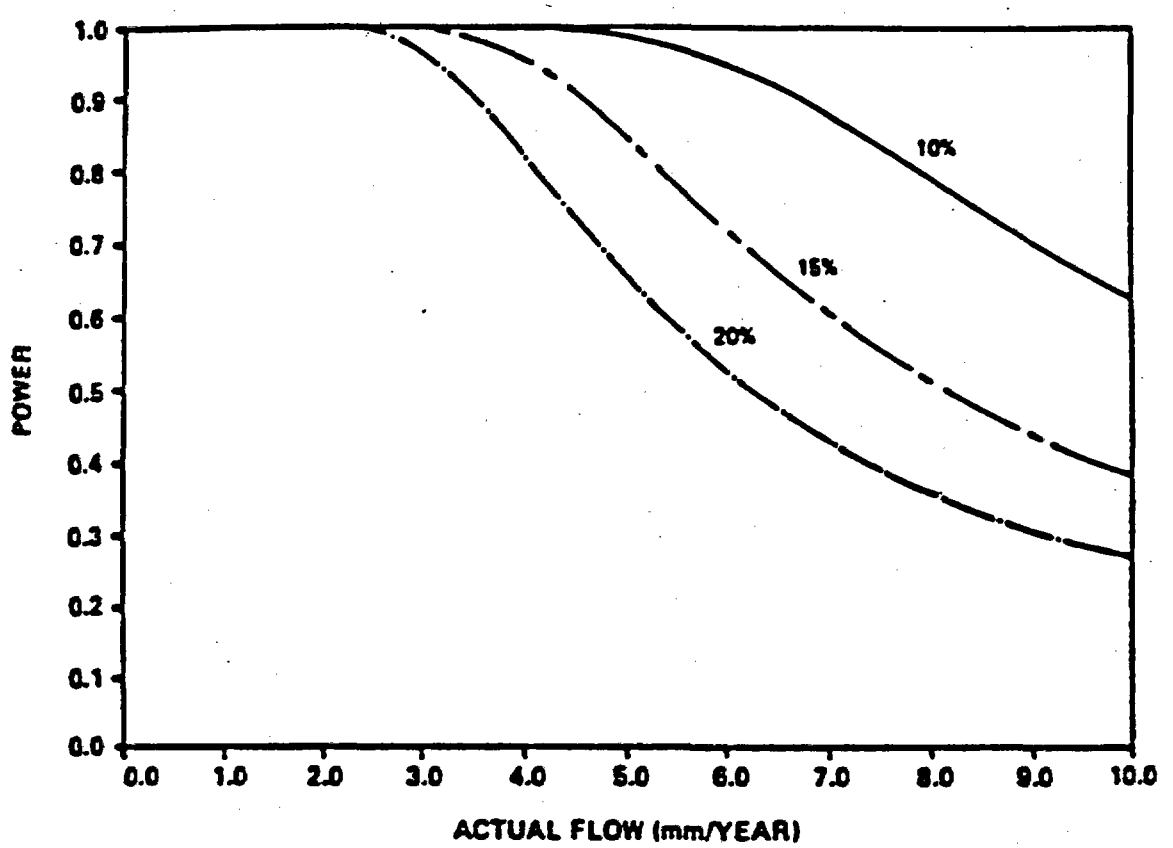

Figure 3. Power curves for measurements of the chlorine-36/total chlorine ratio. The curves are labeled with the $\pm 1\sigma$ values (in percentages) of the individual data points.

The curves in Figure 3 show the probability that the measured data can distinguish between an actual velocity, shown on the abscissa, and the selected hypothetical velocity (greater than or equal to 15 mm/yr in this case). The curves show this probability for different uncertainties associated with the measured data. As the actual velocity approaches the hypothetical velocity, it becomes more likely that the two velocities cannot be distinguished. For example, when the actual velocity is 10 mm/yr, there is a 36% chance that the data will indicate a velocity greater than or equal to 15 mm/yr. However, when the actual velocity is less than about 5 mm/yr, there is essentially no chance that the data will indicate a velocity greater than 15 mm/yr.

Figure 4 shows a plot of the expected half-width of a symmetric 95% confidence interval for the average velocity. This plot shows that a velocity of 1 mm/yr can be estimated within a few percent. However, for a velocity of 5 mm/yr the error associated with the 95% confidence interval will be about ±2 mm/yr with $\pm 1\sigma = 10\%$ measurement errors, which are expected, and will be considerably larger as the measurement errors increase. The maximum flux is expected to be 0.2 mm/yr for a saturation of 0.65 and a porosity of 0.14 (Montazer and Wilson 1984), which is approximately equal to a water velocity of 2 mm/yr. Figure 4 shows that water velocities less than 2 mm/yr will be measured with small uncertainties.

3.5 Equipment and Services Required

Two special facilities are required at the ES-1 site. One is a box at the surface (see Table 2) into which the rubble collected at a particular depth can be poured. The purpose of this box is to separate the sample material from the spoils pile until the sample can be packed in 208-L (55-gal) barrels. The second facility required at the ES-1 site is a metering device (Table 2) to introduce a definite, small quantity of water tracer into all water that is used for mining ES-1. In practice, a bromide tracer will be added to all Well J-13 water used at the exploratory shaft site. A surge tank is likely to be the apparatus of choice for this application. Water samples will be analyzed periodically to verify the water tracer concentration. Water used at the surface for site preparation and dust control also will be tagged with a suitable tracer.

3.6 Representativeness of Velocity Determinations from the Exploratory Shaft

A question can be raised concerning the representativeness of the data from the exploratory shaft when it is extrapolated over the entire area of the repository, because the exploratory shaft allows access to only one point in the area. In the relatively unfaulted portion of Yucca Mountain, spatial results are expected to be fairly uniform. If downward water movement tends to be episodic with either short or long periodicity, interpretation of chlorine-36/chlorine ratios at different depths may be complicated. However, Montazer and Wilson (1984) indicate that such pulses are likely to occur only in the shallow unsaturated zone.

Representativeness of results is also influenced by the degree to which water transport occurs in the matrix of the tuff versus fractures. If most recharge occurs through major structural features as suggested in the conceptual model of Montazer and Wilson (1984), spatial variability may be large. Washes and other areas underlain by structural features would be likely to provide higher velocities, whereas relatively nonfaulted areas would provide lower velocities. The data of most concern to predictions of repository performance are those from the repository level and below. The potential variability in water velocities is therefore unlikely to cause significant changes in the determination of water movement by chlorine-36 tracer studies because the effects of temporal changes in infiltration are likely to be manifested only at shallow depths.

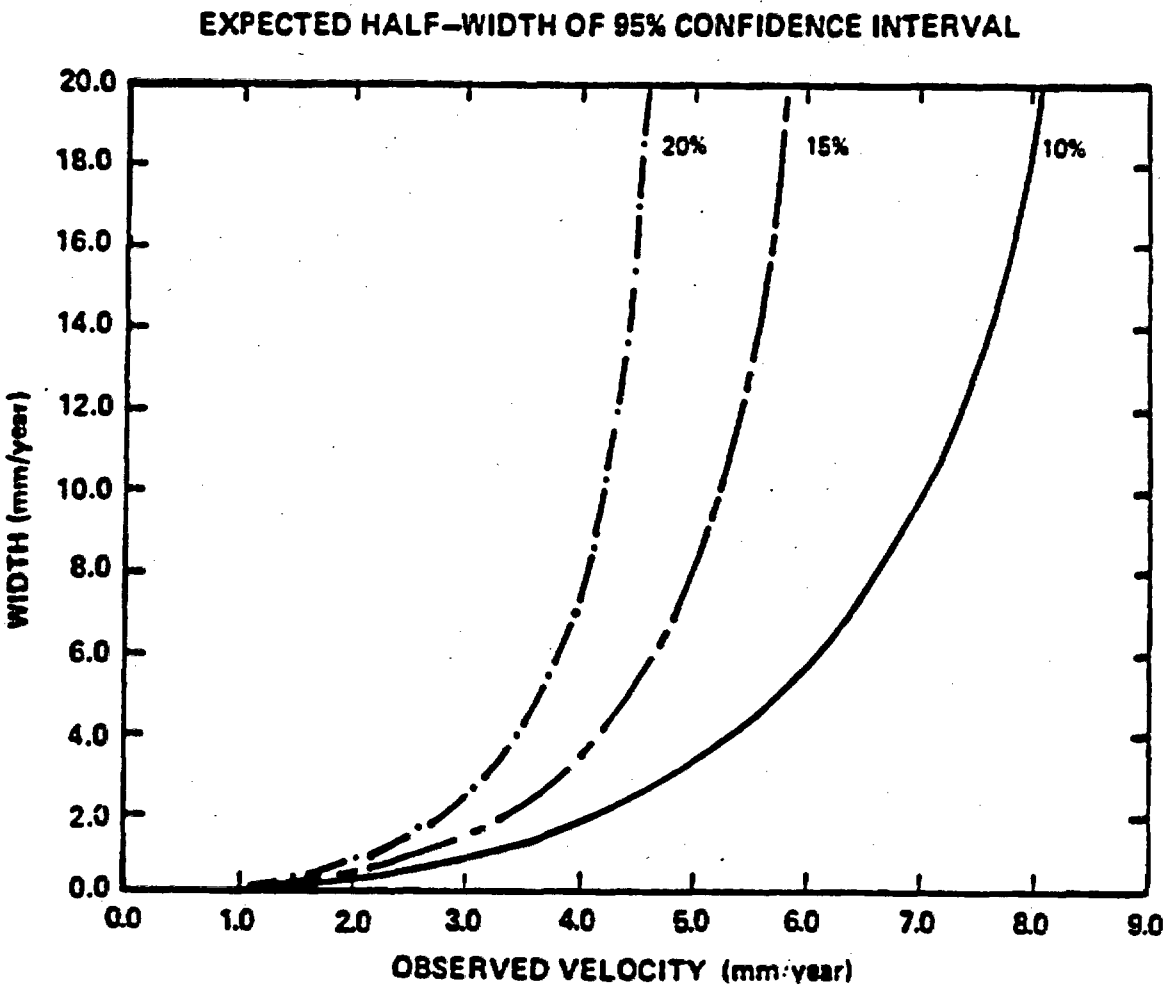

Figure 4. Confidence intervals for measurements of the chlorine-36/total chlorine ratio. The curves are labeled with the $\pm 1\sigma$ values (in percentages) of the individual data points.

TABLE 2

INSTRUMENTATION, EQUIPMENT, MATERIALS, AND SERVICES FOR THIS TEST

Item	Quantity	Description	Procurement Method
1. Box for rubble	1	Nonstandard	Designed and constructed by tester's organization
2. Water tracer metering system	1	Nonstandard	Designed and constructed by tester's organization
3. Water tracer, NaBr	1,120 kg	Standard	Tester's organization purchase order
4. 208-L (55-gal.) barrels	≤60	Standard	Tester's organization purchase order
SERVICES LIST			
1. Depth of designated blast-in ground		Standard	Shift-sinking subcontractor
2. Geologist to select and pack rubble pieces for chlorine-36 analysis (about 40 hours total)			Fenix and Scisson
3. Extract chloride from rubble; analyze for chlorine-36, chloride, and bromide; interpret data		Nonstandard	Tester's organization subcontractor

Samples could be collected at other locations by using a new dry-drilling technique, reverse vacuum drilling (Whitfield 1985). Correlation of the chlorine-36 results with detailed stratigraphic mapping of fracture frequency data would not be possible to the extent that will be possible in the exploratory shaft. If spatially distributed data are likely to improve knowledge of the hydrologic flow through the unsaturated zone, this alternative method of sample collection could be used at the later date.

Another source of material for chlorine-36 analyses is perched water that might be encountered during the construction of the exploratory shaft. If the chloride content of the perched water is 10 ppm, as in Well J-13 water, then 2 to 3 L per sample would be sufficient to obtain material for a chlorine-36 analysis. The data from these analyses might provide information about the rate of water flow in the unsaturated zone at Yucca Mountain.

3.7 Quality Assurance Requirements

The activities in this study plan have been assigned as Quality Level I in accordance with LANL QA procedure TWS-MSTQA-QP-18. (This procedure is currently being revised and

will be issued as TWS-QAS-QP-03.4.) These data may be used in the license application in assessing ground water travel times and ground water flow rates which have a direct bearing on site assessments concerning waste isolation to be used in the license application. The applicable criteria from NQA-1 that apply to this study are shown in Appendix A, along with the procedures and other documents that will satisfy these criteria.

Technical procedures for the work in this study are shown in Table 3.

TABLE 3

TECHNICAL PROCEDURES FOR STUDY PLAN 8.3.1.2.2.2

Activity	Technical Procedure	Type	Anticipated Availability Date
Sampling in Exploratory Shaft	YMP-AP-6.2Q, Borehole sample-handling activities	Nonstandard	TBD*
	YMP-AP-6.3Q, Sample management facility	Nonstandard	TBD*
	YMP-AP-6.6Q, Specimen removal	Nonstandard	TBD*
Field Sampling	To be prepared	Specialized standard	6/90
Sample Crushing and Leaching	To be prepared	Nonstandard	3/90
Chloride Concentration Measurement	ASTM Mercuric Nitrate Titration	Standard	1981
Bromide Concentration Measurement	To be prepared	Specialized standard	3/90
Chlorine Isotope Analyses	To be prepared	Specialized standard	6/90

* Procedures will be ready 30 to 60 days before tests.

4.0 APPLICATION OF RESULTS

4.1 Site Investigation

The work in this study plan will provide information for determining the ground water travel time in the unsaturated zone at Yucca Mountain. Water movements will be

characterized through measurements of the chloride concentrations and the chlorine-36/chlorine ratios, both as a function of depth below surface. Results from this study will be used in assessing water movement at the site. Information from chlorine-36 data may be used for inferring rates of fracture flow relative to matrix flow. The chlorine-36 vertical distribution may permit assessment of the role of convective water movement relative to dispersive movement. This study will be performed in parallel with USGS hydrochemistry studies (8.3.1.2.2.7) and will precede USGS in situ tests (8.3.1.2.2.4). The groups working on these tests plan to exchange information as they progress. The data will be used to validate conceptual models of hydrologic flow in the unsaturated zone by showing, from the chlorine-36 data, whether flow in the unsaturated zone has taken at least 50,000 years. Plans for integrated modeling of the unsaturated zone are described in Sections 8.3.1.2.2.8 and 8.3.1.2.2.9 of the SCP and will be detailed in later study plans. The use of rates of chlorine-36 migration as an upper bound on technetium-99 migration may also provide valuable confirmatory support to results from geochemical studies summarized in Sections 8.3.1.3 and 8.3.5.13 of the SCP.

4.2 Resolution of Performance Issues

The application of results in the site investigation work can be tied directly to resolution of key performance assessment issues. The assessment of total system performance summarized in Section 8.3.5.13 of the SCP is dependent upon the ranges of potential flux passing through and below the repository level. An independent confirmation of flux estimates by rates of movement derived from chlorine-36 studies would provide valuable support to the transport and release predictions. Both containment by the waste package (SCP Section 8.3.5.9) and release from the engineered barrier system (SCP Section 8.3.5.10) are strongly dependent upon the rate of water movement through the repository horizon. Therefore, confirmation of low flux values by independent analytical studies will reduce uncertainties in meeting the waste package and engineered barrier system performance objectives.

As indicated in Section 2 of this study plan, confidence in meeting the 1,000-yr pre-waste-emplacement travel time requirement will increase if chlorine-36 samples below the repository horizon indicate rates of movement far too slow for a shorter travel time.

5.0 SCHEDULE AND MILESTONES

Item	Date
Develop water metering system for NaBr tracer	Constructed and operational by time of exploratory shaft site preparation
Commence sample collection	Start of exploratory shaft construction
Conclude sample collection	Completion of exploratory shaft to depth
Complete chlorine-36/chlorine ratio analyses	1 yr after completion of exploratory shaft to depth
Complete data interpretation and final report	21 months after exploratory shaft completed to depth

A chart showing the anticipated progress in this study is shown in Figure 5.

Milestone	
R495	Infiltration rates from chlorine distributions at Yucca Mountain
M623	Final ESF chlorine-36 report

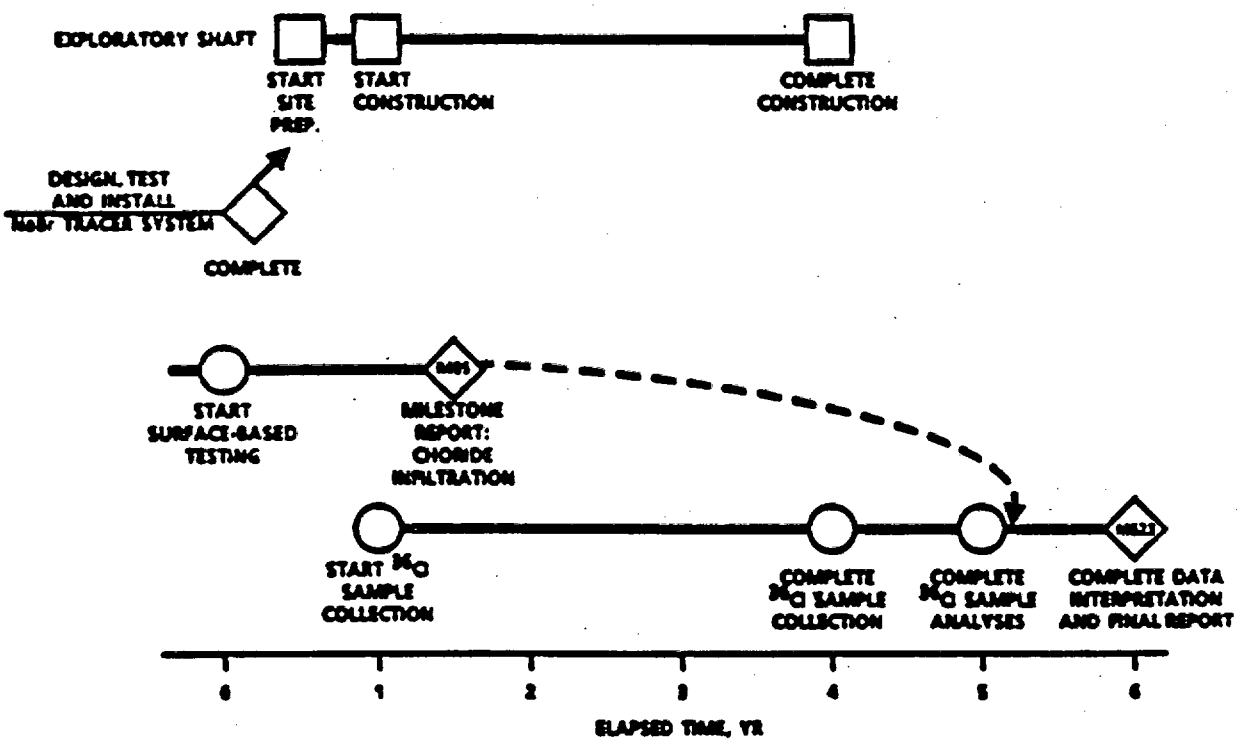

Figure 5. Anticipated Progress in this Study

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APPENDIX A

QUALITY ASSURANCE SUPPORT DOCUMENTATION

Table A-1 lists the applicable NQA-1 criteria for this study and explains how they will be satisfied.

TABLE A-1

APPLICABLE NQA-1 CRITERIA FOR SCP STUDY 8.3.1.2.2
AND HOW THEY WILL BE SATISFIED

NQA-1 Criterion	Documents Addressing These Requirements		Anticipated Date of Issue
1. Organization	The organization of the Office of Civilian Radioactive Waste Management (OCRWM) program is described in Section 8.6 of the SCP. The LANL QA program is described in the LANL-YMP-QAPP and includes a program description addressing each of the NQA-1 criteria. The LANL QA program contains quality administrative procedures (QP) further defining the program requirements.		
	TWS-QAS-QP-01.1	Interface Control	1/31/89
	TWS-QAS-QP-01.2	Stop Work Control	1/31/89
2. QA Program	The LANL QA program is described in the LANL-YMP-QAPP and includes a program description addressing each of the NQA-1 criteria. An overall description of the YMP QA program for site characterization activities is described in Section 8.6 of the SCP.		
	TWS-QAS-QP-02.1	Personnel Certification	1/31/89
	TWS-QAS-QP-02.2	Personnel Training	1/31/89
	TWS-QAS-QP-02.3	Conflict Resolution	1/31/89
	TWS-QAS-QP-02.4	Management Assessment	4/30/89
3. Design and Scientific Investigation Control	This study is a scientific investigation. The following QPs apply:		
	TWS-QAS-QP-03.1	Software QA Plan	4/30/89
	TWS-QAS-QP-03.2	Technical Review	4/30/89
	TWS-QAS-QP-03.3	Scientific Investigation Planning	4/30/89
	TWS-QAS-QP-03.4	Quality Assurance Level Assignment	4/30/89
	TWS-QAS-QP-03.5	R&D Control (Notebooks)	4/30/89
	TWS-QAS-QP-03.6	Design Interface Control	1/31/89
	TWS-QAS-QP-03.7	Peer Review	4/30/89
	TWS-QAS-QP-03.8	Readiness Review	1/31/89
	TWS-QAS-QP-03.9	Scientific Analysis Control	4/30/89

TABLE A-1

APPLICABLE NQA-1 CRITERIA FOR SCP STUDY 8.3.1.2.2
AND HOW THEY WILL BE SATISFIED
(continued)

NQA-1 Criterion	Documents Addressing These Requirements		Anticipated Date of Issue
	TWS-QAS-QP-03.10	Software Documentation and Review	4/30/89
	TWS-QAS-QP-03.11	Software Configuration Management	4/30/89
	TWS-QAS-QP-03.12	Scientific and Engineering Software	4/30/89
	TWS-QAS-QP-03.13	Auxiliary, Commercial and Utility Software	4/30/89
4. Procurement Document Control	TWS-QAS-QP-04.1	Procurement	12/14/88
	TWS-QAS-QP-04.2	Acceptance of Procured Services	1/31/89
	TWS-QAS-QP-04.3	Qualification of Suppliers	1/31/89
5. Instructions, Procedures, and Drawings	TWS-QAS-QP-05.1	Preparation of QPs	12/14/88
	TWS-QAS-QP-05.2	Preparation of DPs	12/14/88
6. Document Control	TWS-QAS-QP-06.1	Controlled Document Distribution	1/31/89
7. Control of Purchased Material, Equipment, and Services	Applicable parts of this criterion are covered in Item 4 (see above).		
8. Identification and Control of Materials, Parts and Samples	TWS-QAS-QP-08.1	Control of Samples	4/30/89
	TWS-QAS-QP-08.2	Control of Data	4/30/89
9. Control of Special Processes	This criterion has been determined to be inapplicable to the scope of work of the LANL YMP.		
10. Inspection	This criterion has been determined to be inapplicable to the scope of work of the LANL YMP.		
11. Test Control	This criterion has been determined to be inapplicable to the scope of work of the LANL YMP.		
12. Control of Measuring and Test Equipment	The control of instrument calibration and data collection is described in the technical procedures referenced in Sections 3.1 through 3.3 of this plan. The following QPs also apply:		

TABLE A-1

APPLICABLE NQA-1 CRITERIA FOR SCP STUDY 8.3.1.2.2
AND HOW THEY WILL BE SATISFIED
(concluded)

NQA-1 Criterion	Documents Addressing These Requirements		Anticipated Date of Issue
	TWS-QAS-QP-12.1	Measuring and Test Equipment	4/30/89
	TWS-QAS-QP-12.2	Control of User Calibrated Equipment	4/30/89
13. Handling, Storage and Shipping	TWS-QAS-QP-13.1	Handling, Shipping, and Storage	4/30/89
14. Inspection, Test and Operating Status	This criterion has been determined to be inapplicable to the scope of work of the LANL YMP.		
15. Nonconforming Materials, Parts or Components	TWS-QAS-QP-15.1	Nonconformances	12/14/88
16. Corrective Action	TWS-QAS-QP-16.1	Corrective Action Control	1/31/89
	TWS-QAS-QP-16.2	Trending	4/30/89
17. Quality Assurance Records	TWS-QAS-QP-17.1	Resident File	12/14/88
	TWS-QAS-QP-17.2	Records Processing Center	12/14/88
18. Audits	TWS-QAS-QP-18.1	Audits	1/31/89
	TWS-QAS-QP-18.2	Surveys	4/30/89
	TWS-QAS-QP-18.3	Auditor/Lead Audit or Cert.	1/31/89

QUALITY ASSURANCE LEVEL ASSIGNMENT SHEET (QALAS)
AND
QUALITY LEVEL ASSIGNMENT CRITERIA SHEET (QLACS)

SIP No. 86/1.2.3.4.1.2

Rev. 0

Activity Natural Isotope Chemistry

Tasks: Chloride and ^{36}Cl Measurements of Infiltration at Yucca Mountain
^{99}Tc Distribution in Soil Relative to ^{36}Cl
Uranium-Series Disequilibrium Feasibility Study

TABLE A-2

QUALITY ASSURANCE LEVEL ASSIGNMENTS
FOR STUDY PLAN 8.3.1.2.2.2

NNWSI QUALITY ASSURANCE LEVEL ASSIGNMENT			
Items/Activities	QA Level	QA Requirements	Technical Justification
Chloride and ^{36}Cl Measurements of	I	1,2,3,4,5,6,7,8, 10,11,12,13,15,	NNWSI SPO-02-02: para 5.2.1.b. & d. (activity will provide site
Infiltration at Yucca Mountain		16,17,18. See attached QLACS	characterization and license application data) and Step 2 of
			QA Level Assignment Checklist

APPROVALS (Signature and Date)

PI A. E. Norris Sept. 3, 1986 TPO D. Gill 9.3.86

QAL Suzanne Dey 9/3/86 WMPO(TECH) M Blanchard 9.4.86

QAIM Paul Liebendorfer 9/5/86 WMPO(PQM) James Blaylock 9/4/86

PI FINAL REVIEW A. E. Norris October 9, 1986

QUALITY LEVEL ASSIGNMENT CRITERIA SHEET (QLACS)

SIP No. 86/1.2.3.4.1.2.

Rev. 0

Activity Natural Isotope Chemistry

Task: Chloride and ^{36}Cl Measurements of Infiltration at Yucca Mountain

PI: A. E. Norris

QA Criterion	Applies	Does not Apply	Comments
1. QA Organization	x		
2. QA Program	x		
3. Design and Scientific Investigation Control	x		Only scientific investigation requirements apply
4. Procurement Document Control	x		
5. Instructions, Procedures, and Drawings	x		
6. Document Control	x		
7. Control of Purchased Material, Equipment, and Services	x		
8. ID and Control of Materials, Parts, Components & Samples	x		

QA Criterion	Applies	Does not Apply	Comments
9. Control of Processes		x	Activities performed under this WBS are not considered to be special processes as defined in Appendix A SOP-02-01
10. Inspection	x		Applicable for surveillance requirements only
11. Test and Experiment/ Research Control	x		
12. Control of Measuring and Test Equipment	x		
13. Handling, Shipping, and Storage	x		
14. Inspection, Test, and Operating Status		x	No hardware generated in this task
15. Control of Nonconformances	x		
16. Corrective Action	x		
17. QA Records	x		
18 QA Audits	x		

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