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YUCCA MOUNTAIN SITE CHARACTERIZATION PROJECT
STUDY PLAN APPROVAL FORM

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WATER MOVEMENT TEST

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ABSTRACT

The water movement test is designed to produce information derived from isotopic measurements of soil, rock and water samples collected from the vicinity of Yucca Mountain that is pertinent for assessing the performance of a nuclear waste repository. Measurements of chlorine isotopic distributions will help characterize the infiltration of precipitation into the unsaturated zone. In the unsaturated zone, natural ^{36}Cl occurs from atmospheric fallout of this radionuclide produced by cosmic-ray secondaries interacting with argon isotopes. When chloride at the surface is carried underground by infiltration, the radioactive decay of ^{36}Cl in the chloride can be used to time the rate of water movement in the deep subsurface. The ^{36}Cl half-life of 301,000 years permits the detection of water movement in the range of 50,000 to 2 million years. Chlorine-36 also occurs as global fallout from high-yield nuclear weapons tests conducted primarily at the Pacific Proving Grounds between 1952 and 1958, at concentrations up to two orders of magnitude above natural background. This latter signal is easily recognizable and can be used to estimate the rate of water movement in shallow soils, providing a means to estimate the upper limit for the present-day infiltration rate. These data are part of the input for developing and evaluating the validity of numerical models of ground-water movement at the candidate repository.

This study consists of a single activity, "Chloride and Chlorine-36 Measurements of Percolation Rates at Yucca Mountain."

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1.0 PURPOSE AND OBJECTIVES OF STUDY

1.1 Purpose

Investigators at Los Alamos National Laboratory (LANL) are conducting studies as part of the Yucca Mountain Site Characterization Project (YMP) to evaluate the suitability of Yucca Mountain as the site for a high-level nuclear waste repository and the ability of the mined geologic-disposal system to isolate the waste in compliance with regulatory requirements. This study plan consists of a single activity:

- Activity 8.3.1.2.2.2.1 - Chloride and Chlorine-36 Measurements of Percolation at Yucca Mountain

This plan describes how the mechanism and rate of water movement downward through the unsaturated zone beneath Yucca Mountain will be evaluated using measurements of chloride concentrations and chlorine isotopic compositions in samples of soil, rock and water collected as part of the site characterization program. Present-day infiltration rates and processes will be characterized in surficial soils by measuring the vertical distribution in these soils of anthropogenic (i.e., bomb-pulse) ^{36}Cl deposited from atmospheric testing of nuclear devices, primarily during the period 1952 to 1958. The soil infiltration rate serves as a boundary condition for evaluating deeper percolation. Rates and pathways of water movement in the deeper unsaturated zone will be evaluated by measuring $^{36}Cl/Cl$ ratios in rock and water samples that will be collected from surface-based boreholes and from the Exploratory Studies Facility (ESF). The 301,000-yr half-life of ^{36}Cl is useful for tracing water movements between 50,000 and 2,000,000 years and is most useful between 100,000 and 1,000,000 years. Detection of bomb-pulse ^{36}Cl in deeper samples would indicate zones of rapid water movement, such as along fractures.

The data from this test will be used as part of the information required by 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.

Figure 1 shows the location of this study plan within the framework of the SCP Geohydrology Program (8.3.1.2) and its subprogram, the Site Unsaturated-Zone Hydrology Investigation (8.3.1.2.2). The Water Movement Test described in this study plan is one of nine studies planned to characterize the unsaturated zone beneath Yucca Mountain. The numbers (e.g., 8.3.1.2.2) used throughout this plan refer to specific sections of the Site Characterization Plan (SCP) (DOE, 1988). The SCP

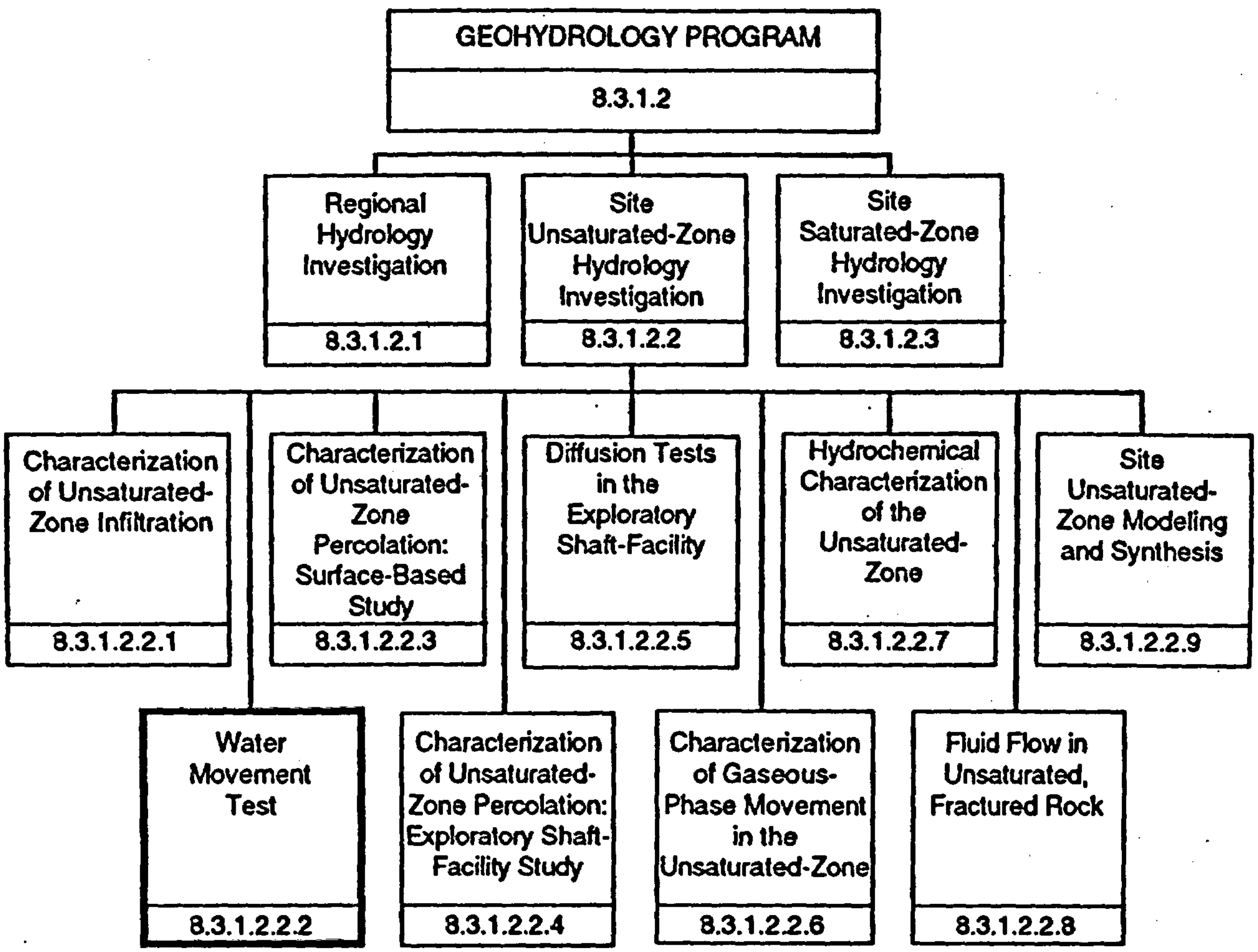

Figure 1. Diagram showing the location of the study plan within the unsaturated-zone investigation, and organization of the geohydrologic characterization program.

describes the technical rationale of the overall site-characterization program and provides general descriptions of the activities described in detail in this and other study plans.

1.2 Resolution of Performance Issues

The rationale for the YMP site characterization program is presented in Section 8.1 of the SCP. The issues-based strategy was guided first by an issue identification procedure 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 key issues and associated information needs that will use the data from this study are those dealing with ground-water travel time and total system performance. The measurements described in this study plan do not directly contribute to these performance issues because the parameters that are used in flow and transport models of Yucca Mountain are generally hydrologic properties of the rocks and fluids. Instead, the isotopic measurements described herein will provide independent corroboration of the results of other studies designed to assess water movement in the unsaturated zone as well as information that may confirm or invalidate model predictions of ground-water velocities and fluxes in the unsaturated zone. Sections 2 and 3 of this study plan discuss the interpretation of the isotopic data with respect to the determination of rates of water movement.

1.2.1 Performance Issue 1.1 (Limiting radionuclide releases to the accessible environment)

Issue 1.1 asks, "Will the mined geologic disposal system meet the system performance objective for limiting radionuclide releases to the accessible environment as required by 10 CFR 60.112 and 40 CFR 191.13?" (SCP Section 8.3.5.13; NRC, 1983; EPA, 1985). This issue requires that the geologic setting, engineered-barrier system, shafts, boreholes and seals be designed to limit the cumulative release of radionuclides for 10,000 years following permanent closure of the repository.

Information Need No. 1.1.1 specifies the site information needed to calculate releases to the accessible environment and establishes tentative goals for performance parameters. One critical parameter is the distribution of fluxes through the unsaturated zone underlying the repository because this zone could provide a significant barrier to liquid flow and hence to radionuclide transport. For scenario class E, the nominal case, a goal of <0.5 mm/yr is set (SCP Table 8.3.5.13-9, p. 8.3.5.13-93). Scenario class C-1 deals with the consequences of local or extensive increases in percolation flux through the unsaturated zone in the vicinity of Yucca Mountain, e.g. as a result of climate changes; a tentative goal is that flux changes will be <0.5 mm/yr with 67% confidence or more (SCP Table 8.3.5.13-12, p. 8.3.5.13-98). The Climate Program

(8.3.1.5) will evaluate this parameter by establishing quantitative confidence bounds on the expected magnitude of change. Although data from the Water Movement Test will not specifically address this parameter, they will nonetheless provide an independent means of estimating infiltration rates and the distribution of fluxes and, thus, complementing other studies addressing present-day infiltration rates and paleo-fluxes through the unsaturated zone.

1.2.2 Performance Issue 1.6 (Ground-water travel time)

Issue 1.6 asks, "Will the site meet the performance objective for pre-waste-emplacement ground-water travel time as required by 10 CFR 60.113?" (SCP section 8.3.5.12; NRC, 1983).

Information Need 1.6.1 identifies site information and design concepts needed to identify the fastest path of likely radionuclide travel and to calculate the ground-water travel time along that path. Performance parameters used by this issue are listed in SCP Table 8.3.5.12-3 (p. 8.3.5.12-28 to 33). One of the parameters needed for establishing initial and boundary conditions is the flux and percolation rate through fault zones, fractures and the rock matrix in the Topopah Spring welded unit in the repository area. A parameter to be used to validate model concepts is isotopic ratios used to estimate ground-water residence times in fractures, fault zones and rock matrices in each unsaturated zone unit below the repository. Data collected by this study will contribute to the estimation of appropriate values and ranges for both of these parameters.

Results obtained for Information Need 1.6.1 are also needed to satisfy other information needs for Performance Issue 1.6: 1.6.2, Calculational models to predict ground-water travel times between the disturbed zone and the accessible environment; 1.6.3, Identification of the paths of likely radionuclide travel from the disturbed zone to the accessible environment and identification of the fastest path; and 1.6.4, Determination of the pre-waste-emplacement ground-water travel time along the fastest path of likely radionuclide travel from the disturbed zone to the accessible environment.

1.2.3 Performance Issue 1.8 (NRC siting criteria)

Postclosure Performance Issue 1.8 addresses NRC siting criteria: "Can the demonstrations for favorable and potentially adverse conditions be made as required by 10 CFR 60.122?" (SCP section 8.3.5.17; NRC, 1983). The Water Movement Test study will provide information relevant to the evaluation of the extent to which several favorable and potentially adverse conditions may be present at the site, as follows:

Favorable condition 1 is that the nature and rates of hydrogeologic and other processes operating within the geologic setting during the Quaternary Period, when projected, would not affect or would favorably affect the ability of the geologic repository to isolate wastes. Data from this study may be used to evaluate paleo-fluxes in the immediate vicinity of Yucca Mountain.

Favorable condition 7 is that pre-waste-emplacement travel time along the fastest path of likely radionuclide travel from the disturbed zone to the accessible environment substantially exceeds 1,000 years. Using an upper bound of 0.5 mm/yr for flux through the repository horizon, the mean travel time would be about 43,400 yr, with a range of 9,500 to 80,200 yr (SCP p. 8.3.5.17-95). The upper range of this time interval is within the range of dating by ^{36}Cl, which may therefore be able to provide confirmatory evidence for the presence of this favorable condition. Alternatively, identification of a component of modern groundwater in the unsaturated units beneath the repository zone would provide evidence that this condition cannot be shown to be present.

Assuming disposal in the unsaturated zone, favorable condition 8, in part, specifies hydrogeologic conditions that include a low moisture flux in the host rock and in the overlying and underlying hydrogeologic units, and/or a laterally extensive low-permeability hydrogeologic unit above the host rock that would inhibit the downward movement of water or divert downward moving water to a location beyond the limits of the underground facility (SCP p. 8.3.5.17-96). The moisture flux at the site is expected to be low, but the actual magnitude of the flux is also expected to vary throughout the host rock and overlying and underlying units because of variations in the matrix and fracture characteristics and structural features such as fault zones. This favorable condition only exists at the site if the moisture flux can be shown to be nearly constant as well as low. In conjunction with other site-characterization studies, this study will provide data useful for establishing the range of variability of moisture fluxes in surface soils and in deeper units. Lateral diversion could occur to some extent, thus reducing the overall downward flux through the repository. The combination of contrasting welded, highly fractured units and nonwelded, porous units with the general gentle dip of the units could promote lateral diversion to some degree. The nonwelded, highly porous unit of the Paintbrush Tuff overlies the welded, highly fractured Topopah Spring unit. Because the matrix permeability of the Topopah Spring unit is much less than the matrix permeability of the nonwelded unit, under partially saturated conditions a permeability barrier is expected at the contact. Capillary barriers to downward movement of water may occur where finer material overlies a coarser material; such may be the case, for example, where the Tiva Canyon welded tuff unit overlies the non-welded tuff of the Paintbrush unit. A capillary barrier may also be present at the base of the Paintbrush hydrogeological unit because the pores of this unit are much smaller than the fractures of the Topopah Spring unit. These various barriers, together with the dip of the beds, could result in a general, eastward lateral diversion. The sampling procedure for this test is designed to detect stratigraphically influenced changes in the rate of water movement through the unsaturated zone. Such changes, if detected, would be useful for establishing limits on the extent to which contacts between units serve as permeability or capillary barriers.

Potentially adverse condition 5 is, in part, the potential for changes in hydrologic conditions that would affect the migration of radionuclides to the accessible environment, such as changes in average interstitial velocity and natural recharge. Potentially adverse condition 6 is the potential for changes in hydrologic conditions resulting from reasonably foreseeable climatic changes. In both cases, one of the scenario concerns to be evaluated is that climatic change causes an increase in infiltration over the controlled area. The performance parameter is the expected flux change attributable to projected climatic changes over the next 10,000 years, with the tentative parameter goal being that the expected flux change will be <0.5 mm/yr. This study addresses this parameter indirectly, insofar as the data may be used to evaluate paleo-fluxes in the immediate vicinity of Yucca Mountain.

1.2.4 Performance Issue 1.9 (Higher level findings)

Issue 1.9 addresses the DOE postclosure siting guidelines and evaluations of repository performance over the next 100,000 yr: "(a) can the higher-level findings required by 10 CFR Part 960 be made for the qualifying condition of the postclosure system guideline and the disqualifying and qualifying conditions of the technical guidelines for geohydrology, geochemistry, rock characteristics, climate changes, erosion, dissolution, tectonics, and human interference; and (b) can the comparative evaluations required by 10 CFR 960.3-1-5 be made?" (SCP Section 8.3.5.18; DOE, 1984).

For resolution of issue 1.9(a), it is noted that three of the postclosure system and technical guidelines listed in 10 CFR 960 include requirements for the geologic setting with respect to the pre-waste-emplacement ground-water travel time, which is addressed by performance issues 1.1 and 1.6. These include the postclosure system guideline (10 CFR 960.4-1), and the technical guidelines for geohydrology (10 CFR 960.4-2-1), rock characteristics (10 CFR 960.4-2-3(a)), and climatic changes (10 CFR 960.4-2-4).

Issue 1.9(b) requires an assessment of the radionuclide releases to the accessible environment under expected conditions for the next 100,000 yr. One evaluation is to emphasize the performance of natural barriers; the other is to emphasize the performance of the total system. This sub-issue will be resolved using the same techniques and system models used to resolve issues 1.1 and 1.6, which address system performance over 10,000 yr and ground-water travel time, respectively.

1.3 Additional Regulatory Justification

The previous section discusses how measurements from this study will provide supporting information required to evaluate compliance with parts of three federal regulations, 10 CFR 60 (NRC, 1983), 10 CFR 960 (DOE, 1984), and 40 CFR 191 (EPA, 1985), insofar as part of these regulations are cited by the performance issues. In addition, this task supports the DOE position required by 10 CFR 60.101(a)(2):

> "...Proof of the future performance of engineered barrier systems and the geologic setting over time periods of many hundreds or many thousands of years is not to be had in the ordinary sense of the word. For such long-term objectives and criteria, what is required is reasonable assurance, making allowance for the time period, hazards, and uncertainties involved, that the outcome will be in conformance with those objectives and criteria. *Demonstration of compliance with such objectives and criteria will involve the use of data from accelerated tests and predictive models that are supported by such measures as field and laboratory tests, monitoring data and natural analog studies.*" (italics added by author).

2.0 RATIONALE FOR STUDY

An overview and justification of the overall study are provided in Section 2.1, including the conceptual model for flow through the unsaturated zone beneath Yucca Mountain, estimates of the rates of water movement in the unsaturated zone, and how information from this study may be used to evaluate the validity of specific hypotheses about hydrologic processes that are included in the conceptual model. Constraints on the study are discussed in Section 2.2. Section 3 of this plan provides additional technical details for specific tests, analyses and methods to be used in the study.

2.1 Technical Rationale

2.1.1 Conceptual model of flow through the unsaturated zone

Generalized conceptual models of moisture movement in the unsaturated zone beneath Yucca Mountain have been developed by Montazer and Wilson (1984) and Sinnock et al. (1986). Wittwer et al. (1992) provides a review of subsequent two-dimensional and three-dimensional site-scale models for the unsaturated zone, highlighting the principal findings of these efforts with respect to flow paths. The major features of these models are summarized by the following set of qualitative hypotheses; details can be found in the original papers and in SCP Sections 3.9.3 and 3.9.4.1. The objectives of the Water Movement Test activity will be to provide data that can be used to establish quantitative bounds on water movement processes and to assess the validity of these hypotheses, as described below in sections 2.1.2 and 2.1.3, respectively.

a) Of the precipitation falling on Yucca Mountain (~150 mm/yr), a small amount (probably <1% of precipitation on the average; see section 2.1.2.1) escapes evapotranspiration and moves into the unsaturated zone to become net infiltration. The amount is spatially variable because of the highly variable topography and variations in geologic units outcropping at the surface, and also temporally variable because of the highly sporadic nature of precipitation. Localized high-recharge rates that greatly exceed the areal average rate are likely in areas of concentrated runoff (e.g., Lehman, 1992).

b) If the influx of infiltration following a given precipitation event is large, e.g. in areas of concentrated runoff, then it may occur as fracture flow in the upper welded unit, the Tiva Canyon, all the way to the base of this unit at the Paintbrush nonwelded unit. If the influx from an event is small, then what is initially fracture flow may get taken up into the matrix of the Tiva Canyon unit by capillary forces and become matrix flow before it reaches the nonwelded unit.

c) At the contact between the welded tuff of the Tiva Canyon unit and the underlying nonwelded Paintbrush unit, lateral flow may occur because of possible capillary or permeability barriers occurring between these two units. Lateral flow may also occur within the nonwelded units because of the significant difference between vertical and horizontal hydraulic conductivities.

d) Flow in the Topopah Spring welded unit is expected to be essentially vertical although it is possible that eastward flow down-dip in the Topopah Spring may occur with infiltration from the Solitario Canyon fault zone on the west side of the repository block. Under steady-state conditions, flow occurs within the matrix for fluxes less than some critical value related to the saturated matrix hydraulic conductivity and capillary pressure characteristics, and occurs predominantly as fracture flow at fluxes higher than the critical value (Nitao et al., 1992).

e) Lateral flow may occur in the Topopah Spring welded unit at its contact with the underlying Calico Hills nonwelded unit, depending upon the flux and upon the particular facies of the Calico Hills. At low fluxes, lateral flow may occur because of capillary-barrier effects within the welded unit where it overlies the higher-conductivity vitric facies of the Calico Hills unit. At high fluxes, vertical as well as lateral flow may result where water moving through fractures in the Topopah Spring unit encounters the low-conductivity zeolitic facies of the Calico Hills unit.

f) The nature of flow in the Calico Hills nonwelded unit is not known. Flow in this unit may be predominantly through fractures, as suggested by observations at Rainier Mesa where fracture flow occurs in nonwelded tuffs with matrix permeabilities 10 to 100 times greater than those of the Calico Hills.

g) At structurally favorable locations, perched water bodies may occur where, for example, offset of units has allowed a lower permeability to dam the unsaturated zone water. Such a water body may or may not be a temporary feature.

h) Flow may occur down the faults, which may even be major conduits for flow.

i) All of these kinds of processes may serve to redistribute the flux within the unsaturated zone, such that the flux becomes significantly spatially variable in this zone.

j) Moisture flow in the unsaturated zone may occur under steady state conditions, predominating as liquid water flow moving vertically downward within the rock matrix, with possible water-vapor movement within the air-filled pores and fractures. Significant liquid-water movement in fractures occurs primarily as episodic, nonequilibrium events that are followed by eventual uptake by the rock matrix of water descending through the fractures.

2.1.2 Estimates of water movement rates

2.1.2.1 Estimates of percolation rates based on hydrologic data

Determining the rate of water movement through the unsaturated zone at Yucca Mountain is one of the most critical tasks for assessing the future performance of a nuclear waste repository, but it is a complex task because of the extremely low fluxes, the great thickness of the unsaturated zone, and the heterogeneity of the tuff units. Water movement in the unsaturated zone occurs as infiltration, percolation, and recharge (Montazer and Wilson, 1984).

- *Infiltration* is the entry of water into soil or rock below the interface with the atmosphere.
- *Net infiltration* refers to water that does not remain in shallow storage and is not rapidly returned to the atmosphere by evapotransporation or shallow lateral flow to washes.
- *Percolation* refers to the flow of water in both the unsaturated and saturated zones.
- *Recharge* is the entry of water into the saturated zone from the unsaturated zone.

In the thick unsaturated zone at Yucca Mountain, the three quantities--net infiltration, percolation, recharge--may be expected to have substantially different values. For example, the present rate of net infiltration is probably somewhat less than the present rate of recharge because groundwater travel time through the unsaturated zone is likely to be on the order of tens of thousands of years and because precipitation appears to have decreased regionally (SCP p. 3-203). However, the present net infiltration may be an indication of expected future recharge. Similarly, percolation rates in individual tuff units do not necessarily reflect net infiltration or recharge because of the possible role of capillary barriers and lateral flow (Montazer and Wilson, 1984).

Direct measurements of infiltration and recharge at Yucca Mountain have not yet been made, but a variety of indirect approaches have been taken which are reviewed by Montazer and Wilson (1984) and Wilson (1985). These estimates are summarized in Table 1. Based on regional water-budget analyses, a range of recharge rates from 0.5 to 4.5 mm/yr was estimated. Using Darcy's equation, estimates of downward percolation rates through the moderately to densely welded portion of the Topopah Spring unit, the host rock for the potential respository, range from 1 x 10^{-7} to 0.2 mm/yr. These estimates were based on hydraulic gradient and effective permeability

Table 1. Summary of recharge and percolation estimates for Yucca Mountain*

Recharge to water table, based on extension of results from regional studies surrounding Yucca Mountain:	
Assume 3% of precipitation becomes recharge	4.5 mm/yr
Recharge through alluvium in Yucca Flat	0.5 mm/yr
Pore velocity at Frenchman Flat	1 mm/yr
Regional water-budget studies	0.5 to 4.5 mm/yr
Percolation flux in Topopah Spring welded unit, based on:	
Geothermal gradient	-1.5 mm/yr
Geothermal gradient, using data from USW UZ-1 after 90 days of operation	-1 to -2 mm/yr
Applying Darcy's equation to data from USW H-1 and USW UZ-1	0.003 to 0.2 mm/yr
Applying Darcy's equation to data from USW G-1 and USW UZ-1	10^{-7} to 10^{-4} mm/yr
Matric-potential distribution in USW UZ-1 (Montazer et al., 1985)	0.1 to 0.5 mm/yr
Geothermal gradient in USW UZ-1 after two years of operation (Montazer et al., 1985)	-0.025 to -0.05 mm/yr
Percolation flux in Paintbrush nonwelded unit, based on unit hydraulic gradient and:	
Geometric mean of hydraulic conductivities	99 to 100 mm/yr
Harmonic mean of hydraulic conductivities	0.012 to 0.12 mm/yr
Percolation flux in Calico Hills nonwelded unit, based on effective hydraulic conductivities and assuming unit hydraulic gradient:	
Vitric facies	55 mm/yr
Zeolitic facies	0.006 mm/yr

* Data from Montazer and Wilson (1984) except as noted otherwise.

data derived from one borehole and from cores recovered from holes at more than one location. Analyses of geothermal heat-flux data from the Topopah Spring unit indicate that the net hydrologic flux may be upward at a rate of 1 to 2 mm/yr because of vapor-phase transport. 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.

2.1.2.2 Estimates of percolation rates to be determined by this study

Understanding the pattern and rate of water movement in the unsaturated zone beneath Yucca Mountain is essential to the site-characterization program because moisture is the expected primary medium for transport of radionuclides from the repository to the accessible environment. Water may be entering the unsaturated zone along ridges, slopes and upland areas through large-aperture fractures following low-intensity, long-duration winter storms and/or high-intensity storms, in addition to water that infiltrates into the washes following rare runoff events. Episodic fracture flow may also occur under washes during rainfall events. Several studies are addressing this issue by various independent approaches: moisture profiling, physical properties, fracture characteristics, climate water-budget modeling. A review and critique of the various techniques commonly used to assess recharge in arid regions is presented in Gee and Hillel (1988). The best indicator of long-term water movement is the spatial distribution of water ages, or residence times, in the subsurface; radiometric methods are the most direct indicators of residence time. Radiometric methods, which are based on measurements of atmospheric tritium, ^{14}C and ^{36}Cl carried underground by percolating water, provide a means for obtaining estimates of residence time as a function of location. These methods also intrinsically average many large-scale heterogeneities. Detection of bomb-pulse levels of these radionuclides provides valuable data on the types of hydrogeologic situations in which episodic fracture flow may be occurring. Finally, measurements of the long-lived radionuclide ^{36}Cl may have inferences for paleohydrology issues such as recharge rates during the pluvial.

The technique chosen to estimate the water travel time through the unsaturated zone in the present study is the measurement of the ratio $^{36}Cl/Cl$ as a function of depth. This ratio will be measured in the soluble chloride fraction of the total chloride of soil and rock samples, i.e. excluding that minor amount of chloride present in mineral structures. 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 ^{36}Cl, which results from cosmic-ray reactions with argon in the atmosphere. The ^{36}Cl half-life of 301,000 yr is appropriate for the travel times calculated from the hydrologic data. If the flux through the Topopah Spring Member is assumed to be 0.2 mm/yr downward, with an average moisture content of 0.1, then estimates of ground-water travel time through this unit exceed 100,000 yr. In model calculations reported by Sinnock et al. (1986), assuming a recharge flux of 0.5 mm/yr, unsaturated-zone travel times from the repository horizon to the water table had a mean of 43,000 years, ranging up to a maximum of 70,000 in the southwest corner of the modeled area where the unsaturated rock mass is thickest. For a flux of 0.1 mm/yr, the mean travel time increased to 213,000 years.

Geochemical properties of chlorine make it a useful tracer of subterranean water movements (Bentley et al., 1986a). Chlorine exists as the nonvolatile chloride anion under most geohydrologic conditions. Chloride ions are among the least sorbed ions on solid surfaces because of their negative charge and small radius in comparison with other common aqueous anions. Several recent studies indicate the validity of using the ^{36}Cl technique for tracing water movements over long times, and it is by far the most successful radionuclide for dating waters older than 100,000 years (Fabryka-Martin et al., 1987; Lehmann et al., 1993). Ground-water residence times and conceptual ground-water flow paths have been inferred from $^{36}Cl/Cl$ measurements for dissolved chloride in groundwater from several aquifers (Bentley et al., 1986a,b; Andrews et al., 1986, 1989; Phillips et al., 1986a; Fabryka-Martin et al., 1991).

At Yucca Mountain, the technique discussed in this study plan will measure the isotopic composition of soluble chloride carried into the subsurface by meteoric water and now present in pore water or on the surfaces of the mineral grains. This chloride traces the movement of water in the liquid phase through the unsaturated zone. Analyses of ^{36}Cl in cuttings from the USW UZ-1 borehole are described in Norris et al. (1990) and show the efficacy of this approach. The cuttings were leached with deionized water to extract soluble chloride, which was then analyzed for its $^{36}Cl/Cl$ ratio. Unambiguous interpretation of these data is not possible for a number of reasons, some of which are identified below (Norris et al., 1990); in addition, the data were not obtained using approved technical procedures or following quality assurance procedures of the caliber required by the YMP. Nonetheless, the data are useful for illustrating the manner in which such data will be interpreted in this study.

The results obtained by Norris et al. (1990) are shown in Figure 2, in which the measured $^{36}Cl/Cl$ ratios are plotted as a function of depth. The ratios are to be compared against the estimated local ratio for modern chlorine, prior to the advent of atmospheric testing of nuclear devices, 5×10^{-13} (Norris et al., 1987). The ratios measured for the depths of 311 and 372 m are considerably less than the modern ratio, indicating the possibility of significant ^{36}Cl decay and hence long residence times. Upper age limits of 340,000 and 720,000 years are calculated for the meteoric chloride associated with these samples, respectively. These residence times correspond to lower bounds on the linear water velocity of 0.9 and 0.5 mm/yr at these depths.

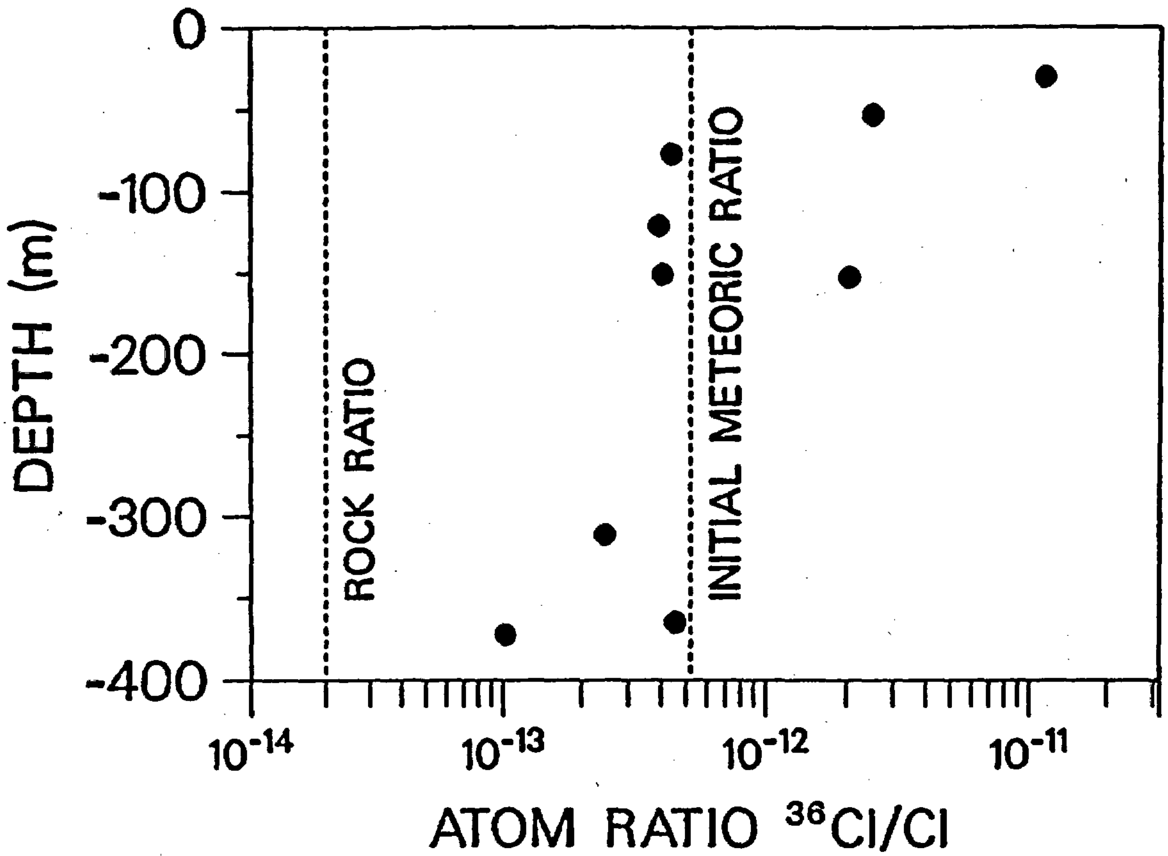

Figure 2. Preliminary results of $^{36}Cl/Cl$ analyses of USW UZ-1 cuttings (Norris et al., 1990)

A firm interpretation of the ratios in terms of water residence time cannot be made, however, because the leached chloride contains some unknown proportion of rock chloride, which serves to dilute the meteoric $^{36}Cl/Cl$ ratio. This limitation is being addressed in future analyses by the use of Cl/Br and stable chloride ratios in the soluble halide fraction of rock samples to indicate the extent of dilution of meteoric chloride by rock chloride (section 2.2). The $^{36}Cl/Cl$ ratio observed at 364 m is statistically the same as the modern value, indicating that water residence time at this depth may be too short to be sensitive to the ^{36}Cl method, i.e., < 50,000 years. However, the isotopic signal for this sample may have been contaminated by drilling fluid lost during the drilling of USW G-1 nearby and unambiguously detected in USW UZ-1 (Norris et al., 1990). Ratios at depths of 30, 52 and 152 m clearly contain a component of bomb-pulse ^{36}Cl, which provides evidence of water movement through fractures (Norris et al., 1990). Additional evidence for fast-path flow through fractures is provided by bomb-pulse levels of ^{36}Cl observed in cutting samples collected in G-Tunnel, 400 m beneath the surface of Rainier Mesa (Norris et al., 1990).

2.1.2.3 Estimates of net infiltration rate to be determined by this study

A second objective of this study plan is to help assess the range of present-day infiltration rates by measuring soil profiles of ^{36}Cl in different geomorphic sites in the immediate vicinity of Yucca Mountain. The question is, how spatially variable is infiltration? If it is 0.1 to 0.5 mm/yr on the average, does it vary from 0 to 50 mm/yr, or up to 1000 mm/yr in certain spots? The mechanism and rate for water movement in the unsaturated zone is very sensitive to the flux on a local scale. Although the number and location of soil profiles planned to be measured in this study will not be sufficient to estimate the total range of infiltration rates, data from this study will nonetheless expand the data base of such rates applicable to Yucca Mountain.

Infiltration rates in arid soils have been evaluated using ^{36}Cl as a water tracer by several researchers in the past few years (Norris et al., 1987; Phillips et al., 1988; Scanlon et al., 1990; Scanlon, 1992; Walker et al., 1991). Only a negligible proportion of the ^{36}Cl measured in these studies was from cosmic-ray reactions with argon in the atmosphere; the isotopic composition of the samples was dominated by bomb-pulse ^{36}Cl that was deposited globally as fallout from high-yield nuclear weapons tests that were primarily conducted at the surface of the Pacific Ocean between 1952 and 1958 (Bentley et al., 1986a; Synal et al., 1990). The estimated source term for bomb-pulse ^{36}Cl is shown in Figure 3 in which it can be seen that peak fallout concentrations exceed background levels by factors of 100 to 1000. Not included in Figure 3 are contributions of ^{36}Cl produced by Chinese and French tests conducted after 1962, with the most recent test being one in 1981. Although minor, these ^{36}Cl sources were nonetheless sufficient to maintain global fallout levels of ^{36}Cl somewhat above natural levels until about 1985 (Synal et al, 1990). Figure 3 also contrasts the ^{36}Cl and tritium

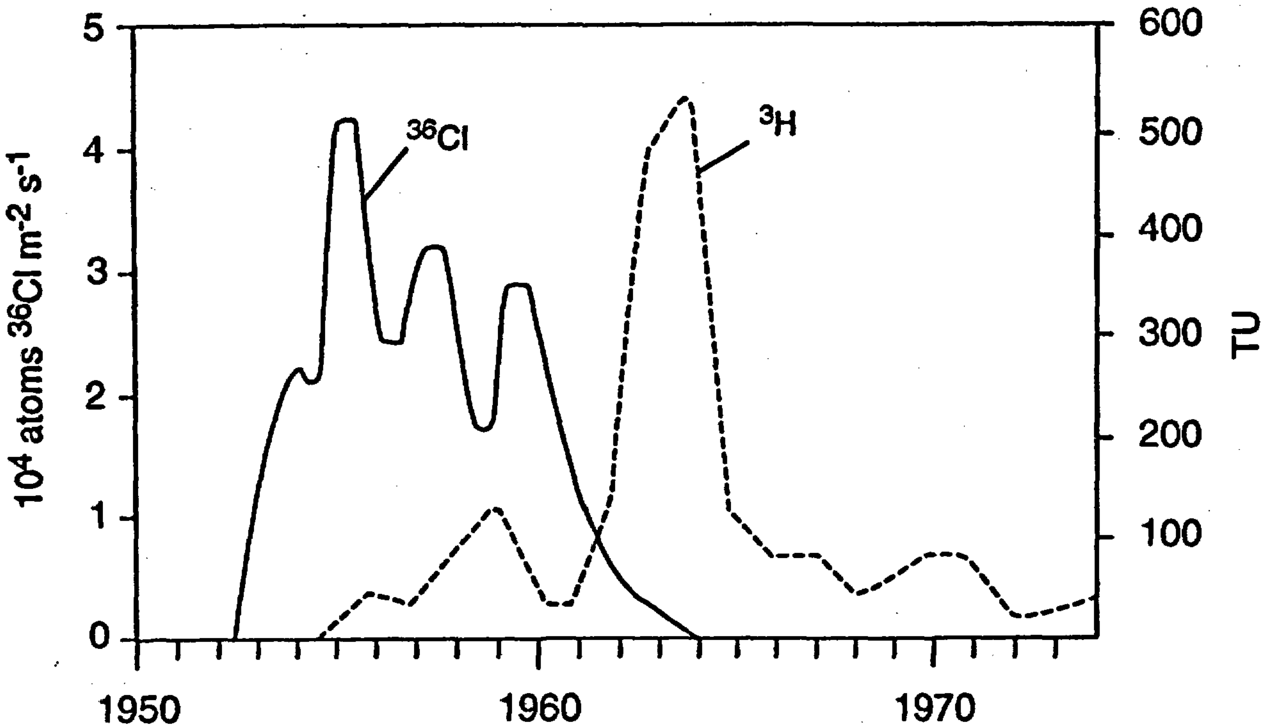

Figure 3. Temporal variations in predicted bomb ^{36}Cl fallout between 30°N and 50°N latitude (Bentley et al., 1986) and in bomb ^{3}H fallout (decay corrected to 1989) for the northern hemisphere (figure adapted from Scanlon, 1992a).

bomb pulses, illustrating the differences in the timing and relative magnitudes of these elevated signals.

Norris et al. (1987) used measurements of bomb-pulse ^{36}Cl 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 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 35 years. This value represents a measurement of flux that is valuable in establishing the upper bound for the amount of water flowing downward through the unsaturated zone.

2.1.3 Role of study in assessing the validity of conceptual model for liquid-water movement in the unsaturated zone

The conceptual model described in Section 2.1.1 consists of a collection of hypotheses about the response of the hydrogeologic system to a given set of boundary conditions and an initial hydrologic state, based upon fundamental concepts about fluid movement and storage. Section 2.1.2.3 described how data from this study could be used to set limits on one of the boundary conditions, i.e. the magnitude of the flux across the surface boundary. This section describes how information from this study could be used to test individual hypotheses incorporated into the conceptual model.

Specific hypotheses and their alternatives which may be amenable to testing using results from this study are listed in Table 2, which is a modified version of Table 8.3.1.2-2 presented in the SCP. For the sake of clarity and convenience, columns from the original source table have been combined and editted in some cases. The purpose of Table 2 is to emphasis aspects in which the present study may serve to discriminate between alternative hypotheses or to reduce uncertainty. This hypothesis-testing table summarizes the following aspects of modeling unsaturated-zone hydrogeology at the site:

- Column 1 - current (DOE, 1988) hypotheses regarding how the site can be modeled and the rationale for these hypotheses
- Column 2 - alternative hypotheses that are also consistent with available data; and
- Column 3 - the need to reduce the uncertainty associated with this current understanding.

Most of the hypotheses deal with the role of fractures and faults in controlling water movement, and with the appropriate modeling approach to use for these features. The hydrologic process of matrix/fracture interaction is one of the most important conceptual issues to be evaluated at the Yucca Mountain site. Preliminary model

Table 2. Current representation and alternative hypotheses for conceptual models of flow in the unsaturated zone (modified as described in the text from SCP Table 8.3.1.2-2a, pp 8.3.1.2-52 to 67). Data collected in this study may be useful for testing the hypotheses shown in this table.

ISSUE AND CURRENT HYPOTHESIS (H)	ALTERNATIVE HYPOTHESIS (A)	NEED TO REDUCE UNCERTAINTY
1. ROLE OF FRACTURES H: Fractures and fracture systems are barriers to or conduits for liquid-water flow, depending on ambient matrix saturation, based on evidence that spontaneous longitudinal flow in fractures is not initiated until matrix is at or near saturation	A: Water may move longitudinally within fractures even at low values of matrix saturation	High: Hydrologic interaction between matrix and fractures will affect possible magnitudes of ground-water travel time and water inflow to underground facility
2. ROLE OF FAULTS IN WELDED TUFF UNITS H: Faults are barriers to or conduits for liquid-water flow in welded tuff units, depending on ambient matrix saturation based on expectation that fault hydraulics in welded units are probably similar to those of fractures	A: Faults are everywhere conduits for liquid-water flow in welded tuffs	High: Faults transect entire UZ; their hydrologic significance needs to be assessed
3. ROLE OF FAULTS IN UNWELDED TUFF UNITS H: Faults are barrier to fluid flow in nonwelded tuff units for all matrix saturations based on expectation that faults in ductile, non-welded units are probably sealed with fault gouge, clays or mineralization	A: Faults are everywhere conduits for liquid-water flow in non-welded tuff units	Med.: Sealed faults could produce temporary perched water bodies under transient conditions
4. NATURE OF WATER MOVEMENT IN FRACTURES AND FAULTS H: Transient, non-equilibrium flow of water occurs in open fractures and faults, based on evidence indicating the viability of this effect in the Tiva Canyon unit	A: Water is rapidly imbibed into the rock matrix at the fracture boundaries	Med.: This may be the principal mechanism by which water enters the UZ as net infiltration

Table 2 (continued)

ISSUE AND CURRENT HYPOTHESIS (H)	ALTERNATIVE HYPOTHESIS (A)	NEED TO REDUCE UNCERTAINTY
5. HYDROLOGIC INTERACTION BETWEEN PORES AND FRACTURES H: Hydrologically interconnected fracture systems and rock matrix define a macroscopic composite or equivalent porous medium	A: Fractures and fracture systems must be regarded as distinct hydrologic entities	Med.: Site-scale modeling will depend on validity of this hypothesis
6. UPPER BOUNDARY CONDITION H: The land surface is the flux boundary for liquid water, air, and water vapor; flux across this boundary is spatially and temporally variable	A: The land surface may be a zero-or uniform-flux boundary for any or all fluid phases	High: Is net infiltration occurring? Damped and redistributed with depth? Capable of transient penetration into deep UZ?
7. PERCHED WATER STABILITY H: Perched-water bodies and capillary-barriers may be temporarily present in the UZ system based on the assumption that perched water bodies are intrinsically unstable and hence can occur only under transient conditions	A: Localized water inflows to the UZ are sufficient to sustain a perched-water body or capillary barrier in a quasi-stable state	Med.: Perched-water bodies are probably transient phenomena and would disperse if they are formed
8. PORE-WATER SOURCE H: Pore-water chemical and isotopic composition reflects pore-water source regions and mechanisms based on the expectation that these characteristics are determined by degree of pore-water interactions with rock matrix	A: Chemical and isotopic concentrations may be non-diagnostic mixture of waters from different sources	Med.: Isotopic and anionic concentrations could be used as supporting if not definitive evidence for source of UZ water

Table 2 (continued)

ISSUE AND CURRENT HYPOTHESIS (H)	ALTERNATIVE HYPOTHESIS (A)	NEED TO REDUCE UNCERTAINTY
9. APPROPRIATE MODELING APPROACH FOR FRACTURES H: Discrete fractures and fracture networks can be modeled as equivalent porous media, an assumption that is is intuitively plausible although fluid-flow mechanisms in partially saturated media are not well-known or quantified	A: Fluid flow in fractures is inherently dynamic and cannot be treated by simple global models	High: Quantified mechanisms of matrix-fracture interactions and bounding uncertainties need to be known
10. PATHWAYS FOR WATER MOVEMENT H: Liquid-water flow in the Topopah Spring is restricted to the rock matrix because expected fluxes are too low in this unit to induce spontaneous flow in fractures	A: Longitudinal flow of water in fractures may occur locally or episodically over a wide range of rock-matrix saturations	High: Dynamics of matrix-fracture hydrologic interactions need to be elucidated

calculations of flow through the unsaturated zone have often assumed a unit hydraulic gradient (i.e., vertical flow), used average hydraulic conductivities, and assumed the percolation flux is uniformly distributed across the upper boundary of the modeled zone. Under such assumptions, for a flux less than the saturated hydraulic conductivity of the rock, the flow will always be through the matrix, never in the fractures. However, there are several realistic scenarios which could lead to fracture flow. For example, if infiltration were to be modeled as episodic rather than spatially uniform and constant, the flux could exceed the matrix hydraulic conductivity and induce fracture flow. A capillary barrier could act to funnel water toward conductive fractures. Finally, model calculations by Bloomsburg et al. (1989) show that fracture flow could be induced because of the expected heterogeneities in rock properties such as hydraulic conductivity.

The mechanisms of water transport in the unsaturated zone are required to be understood in order to use appropriate models for groundwater travel time and radionuclide transport. It is possible for ^{36}Cl/Cl data collected in this study to shed light on the mechanism of water flow if rapid fracture flow dominates to the extent that bomb-pulse ^{36}Cl is observed at great depths or if the hydrologic flow is so slow that hydrologists would describe the mechanism as porous flow. Consequently, the sampling program for this study will focus to a large extent on identifying which faults and fracture zones may be conduits for flow in the unsaturated zone, and under what hydrologic conditions, and which zones may be barriers to flow.

2.2 Constraints on the Study

2.2.1 Need to simulate repository conditions

Analyses for this study will be performed on samples from Yucca Mountain in the vicinity above, below and within the repository block. Consequently, results will help to describe infiltration processes in the repository area. The extent to which the results represent past, present or future infiltration rates of the repository area depends upon factors particular to each test.

2.2.2 Required accuracy and precision, limits of methods, and capability of analytical methods to support the study

The basic measurements in this test are the determinations of chloride, bromide, ^{36}Cl/Cl and ^{37}Cl/^{35}Cl ratios in soil, rock and water samples from Yucca Mountain. Moisture content and bulk density for soil and rock samples will be determined in this study or obtained from the YMP technical data base, as appropriate. Selected and alternate analytical methods are listed in Table 3. These methods were selected on the basis of their precision and accuracy. The accuracy and precision of each of the

Table 3. Parameters measured in this study

PARAMETER	EXPECTED RANGE OF VALUES	ANALYTICAL PROCEDURE S - Selected A - Alternate	DETECTION LIMIT AND PRECISION (see Note A)	REF.
Bromide	1 ppb - 20 ppm	S - Ion chromatography A - Colorimetry A - Neutron activation A - Ion selective electrode	1 ppb, ± 2-4% for 50 ppb 4 ppb 0.02 ppb 50 ppb, ± 30% for 5 ppm	a b c d
Chloride	0.1 - 1000 ppm	S - Ion chromatography A - Colorimetry A - Ion selective electrode	0.1 ppm, ± 3% for 17 ppm 8 ppm, ± 5% for 10 ppm 2 ppm, ± 20% for 10 ppm	e f g
$^{36}Cl/Cl$	10^{-15} to 10^{-11}	S - Accelerator mass spectrometry A - None available	1×10^{-15}, ± 5% for ratios $> 2 \times 10^{-13}$	h
Shift in $^{37}Cl/^{35}Cl$ ratio relative to seawater chloride (Note B)	-1 to 3 per mil	S - Mass spectrometry A - None available	N.A., ± 0.16 per mil	i
Moisture content	0 - 60 vol %	S - Convection oven A - Microwave oven	To be determined	j k
Bulk density	0.5 - 2 g/cm^3	S - Clod method A - Core method	To be determined	l

N.A. Not applicable
Note A: Detection limits and precision for the selected methods are estimates reported in the literature. In most cases, precision varies as a function of the measured value. The manner in which the precision of each measurement will be determined will be described in the appropriate detailed technical procedure.
Note B: Atomic ratio $^{37}Cl/^{35}Cl$ for seawater chloride is approximately 0.320 (Walker et al., 1989).

References

(a) ASTM, 1990c; Lundstrom et al., 1984; Morrow and Minear, 1984; (b) Fishman and Friedman, 1989a; Whittemore, 1988; (c) Luten et al., 1977; Whittemore, 1988; (d) ASTM, 1990a; Whittemore, 1988; (e) ASTM, 1990c; (f) Fishman and Friedman, 1989b, 1989c, 1989d; ASTM, 1990e; (g) ASTM, 1990e; (h) Sharma et al., 1990; (i) Eastoe et al., 1989; (j) ASTM, 1990b; (k) ASTM, 1990d; (l) Blake, 1965

techniques are discussed in various publications describing the methods and will be defined, as appropriate, in the technical procedures developed for their implementation. The LANL/YMP Quality Assurance Plan requires that each technical procedure address the following aspects of data collection: (1) requirements for precision and accuracy; (2) acceptance and rejection criteria; (3) calibration requirements; and (4) potential sources of error and uncertainty in measurements.

Only one method is available for each of the chlorine isotopic measurements. The specific activity of ^{36}Cl in the environment is so low that measurements of the isotopic abundance of ^{36}Cl are only feasible by tandem accelerator mass spectrometry. The sensitivity of this technique currently is approximately one atom of ^{36}Cl in 10^{15} atoms of chlorine. The background meteoric ratio of $^{36}Cl/Cl$ measured in surficial deposits of alluvium at Yucca Mountain is 5×10^{-13} (Norris et al., 1987), which is more than 100 times greater than the limit of this technique. Any samples collected in which ^{36}Cl has undergone detectable radioactive decay will result in $^{36}Cl/Cl$ ratios lower than that of the cosmogenic background.

Section 3.6 provides an in-depth statistical analysis of the expected precision of the overall results of this study.

2.2.3 Potential impacts of activities on site

Analytical activities described in this study will have a minor impact on the natural-state site conditions, and no adverse effect on the ability of Yucca Mountain to isolate waste. The proposed work should not affect the site in terms of either the ESF or repository design.

2.2.4 Time required vs. time available to complete the study

A tentative schedule of work activities and reports is given for this study in Section 5. The analytical techniques for this study are available and are described in Section 3 of this plan. The start of the activities for collection and analysis of ESF samples and cuttings from surface-based drilling is constrained by the schedule for starting those activities and by the timeliness with which samples generated by those activities are made available to this study. The ^{36}Cl analyses can only be performed when the accelerator mass spectrometer is available, which can require a wait of several months because of competing users or to downtime for the machine. ESF operations are not affected by this wait.

The time required for interpretation of the chlorine isotope data does not constrain any other activity because information provided by this study is not critical for submission of the license application. Although data from this study will support license application through testing and evaluation of unsaturated-zone flow and transport models that will be used by both site characterization and performance assessment, the ^{36}Cl and other data derived from this study represent only one of several approaches being taken within the site-characterization project to provide confidence in the application of these models to Yucca Mountain. Nonetheless, the study is important in that it will provide independent and unique information to address potential site suitability issues related to ground-water flow and to augment the data set available to the modeling of transport in the unsaturated zone.

2.2.5 Potential for interference from other field activities

The interferences to which this study is susceptible from other field activities include those activities involving the use of water, the introduction of chlorine which could alter the in-situ $^{36}Cl/Cl$ ratio, or the introduction of bromide which could alter the in-situ Cl/Br ratio. Such activities include previous or future wet drilling or mining near dry-drilled sites from which cuttings for this study are to be collected, possibly seismic reflection and refraction surveys in which explosives are used, and possibly atmospheric fallout of ^{36}Cl produced by nuclear test activities conducted on the surface of the Nevada Test Site during the 1950's and 1960's. (Chlorine-36 produced during underground testing at the Nevada Test Site will not affect this study). Potential interferences from other YMP field activities are discussed below.

2.2.5.1 Potential sources of water

Water used during drilling and mining could be deleterious to the ^{36}Cl analyses of tuff samples in two ways: the water may cause movement of the natural ^{36}Cl, thus giving a false indicator of flow rate and path, or the water may itself contain chloride and ^{36}Cl, thus modifying the measured $^{36}Cl/Cl$ ratio. The problem of water contact will be minimized by several approaches. First, borehole sample requests will be limited to cuttings collected by dry-drilling techniques. Second, water used during wet-drilling of surface-based holes or during mining of the ESF will be tagged with a tracer. Third, if the ESF is mined by drill and blast techniques, ^{36}Cl analyses of the rubble will be done on samples with some minimum dimension on the order of 5 or more cm (2 or more in) so as to minimize the effects of water on the sample interior. Also, rubble samples will be removed from the vicinity of the working surface prior to the water wash-down that follows a blasting round. Because this sample size restriction may bias the results by not sampling fractured areas, dry-coring methods will be requested in the vicinity of faults and extensive fracture networks encountered during mining of the ESF.

Water used during wet-drilling of previous surface-based holes was sometimes, but not always, tagged with a tracer. If the results obtained by this study are anomalous for any given dry-drilled borehole, then the drilling reports for any nearby wet-drilled holes will be examined to determine the source of water used, the presence or absence of a tracer, and the nature of any other additives which conceivably could lead to a shift in the Cl/Br, $^{37}Cl/^{35}Cl$, and $^{36}Cl/Cl$ ratios.

Another source of water is precipitation during borehole drilling and sample collection. In the event of inclement weather, appropriate precautions will be taken by staff of the Sample Management Facility, which is responsible for sample collection, to protect the integrity of the samples from the time of collection until delivery to the investigator for analysis. The requirement for these precautions will be clearly stated in the appropriate documentation produced by the principal investigator in accordance with DOE adminstrative procedures.

2.2.5.2 Potential sources of chloride and ^{36}Cl

Chlorine-36 was produced in the 1950's and 1960's by activation of chloride in surface soils around atmospheric tests of nuclear devices in Yucca Flat and Frenchman Flat, and by testing of nuclear-powered rocket engines in Area 25. These local sources of high ^{36}Cl concentrations presumedly have been transported well below the surface by the present time, thereby limiting the potential for spreading as airborne contamination. However, drilling equipment used in the affected areas prior to drilling a given borehole for YMP sampling conceivably could lead to contamination of the ream-bit cuttings collected from the YMP borehole for ^{36}Cl analysis. The need to take precautions against this possibility will be specified in the appropriate documentation produced by the principal investigator. In the event of an anomalous ^{36}Cl result, the prior history of drilling equipment used to drill that particular YMP borehole will have to be known.

Other than for the case indicated above, accidental contamination of samples with chloride or ^{36}Cl during field collection of samples is not expected to be a problem. Chlorine-36, unlike ^{14}C and tritium, is not transported in the vapor phase. No special atmospheric protection is necessary during sample collection or storage. Most chlorine that might accidentally contaminate the samples for this study would come from chloride ions in the water used in drilling or construction and from chlorine in explosives if these are used during seismic profiling studies or mining operations. The following precautions will be taken to minimize the potential for contamination of samples:

a) For seismic work, the Principal Investigator will interface with appropriate personnel to identify where such activities are to be conducted and will avoid collecting samples from those locations.

b) For the ESF, in the event that water and/or explosives are used, the selection of larger pieces of rubble and the postponement of the customary washdown after blasting are two steps which would be taken to mitigate potential sample contamination problems.

c) At present, water from Well J-13 is proposed for use in wet-mining and wet-drilling activities. The chloride content of Well J-13 water is approximately 10 mg/L; its $^{36}Cl/Cl$ ratio was previously measured to be $5.3 \pm 0.4 \times 10^{-13}$ although this value would be remeasured if this water were used in drilling or mining operations. This information can be used to evaluate the extent to which contamination of a given sample may have affected the analytical results.

d) Any water used underground will be tagged with a tracer. The tracer content of the rock samples selected from the ESF for ^{36}Cl analysis will be measured to permit the calculation of the chloride that might have been introduced from Well J-13 water. Finally, a chlorine analysis of any 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.

Chlorine-36 is also produced from neutron capture by ^{35}Cl. Any production of ^{36}Cl as a result of neutron moisture monitoring is immaterial because the cuttings samples for analysis are collected during construction of the hole, before installation of the detectors. However, production by natural processes in the rocks must be quantified and taken into account in the interpretation of the ^{36}Cl data. Neutrons are produced in the subsurface as a result of 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. The in-situ ^{36}Cl production rate in a rock can be crudely estimated from its elemental composition using the model described by Feige et al. (1968), using the updated parameter values of Andrews et al. (1989) and Heaton et al. (1990). By this method, $^{36}Cl/Cl$ ratios of chlorine in tuffs at Yucca Mountain are on the order of $2\text{-}4 \times 10^{-14}$. The results of the model are crude because the model assumes homogeneous distributions of the α-emitters and α-targets. Consequently, the production rate is more accurately determined by measuring the $^{36}Cl/Cl$ ratio in rock samples after the meteoric component has first been removed by leaching. Both approaches--model calculations and $^{36}Cl/Cl$ analyses of samples--will be taken in this study. Elemental analyses will be conducted on selected samples in order to provide data for the model calculations. The objectives of the modeling and $^{36}Cl/Cl$ analyses will be to develop a best estimate for the average in-situ $^{36}Cl/Cl$ ratio, to evaluate whether the ratio varies significantly within or between lithologic units, and to assess the magnitude of variability in the ratio.

Measurements which may be used to correct the measured $^{36}Cl/Cl$ ratios for the contribution from rock chloride are Cl/Br ratios and stable chlorine isotope ratios.

These two types of ratios are expected to be significantly different depending on the origin of the chlorine. Cl and Br in the atmosphere are derived predominantly from the ocean, in which the Cl/Br weight ratio is 290. The ratio in inland precipitation is considerably less than this value because Cl and Br are fractionated from one another both during their transfer from the ocean to the atmosphere, and again when they are removed from the atmosphere by dry and wet fallout (Winchester and Duce, 1967). Samples analyzed during the research and development stage of this study indicate that the ratio for meteoric water at Yucca Mountain is about 130, which is in agreement with Cl/Br ratios reported for meteoric water in the literature (Norris et al., 1990). In contrast, Cl/Br ratios in tuff from Yucca Mountain are greater than that of the pure meteoric end-member, with an approximate value of about 500 (Norris et al., 1990). Cl/Br ratios in volcanic rocks are most likely governed by differences in the distribution of volatile halogen compounds between the molten silicate phase and the gas phase (Noble et al., 1967; Yoshida et al., 1971). Both end-member ratios will be thoroughly characterized during this study to develop a best estimate for the average ratios, to evaluate whether the ratios vary spatially, and to assess the magnitude of variability in the ratios.

Stable chlorine isotope ratios are an alternative method which may be used to correct the measured $^{36}Cl/Cl$ ratios. Chloride in the atmosphere is derived predominantly from the ocean, and the seawater $^{37}Cl/^{35}Cl$ ratio has been adopted as the standard against which the stable Cl isotopic ratios of other samples are to be compared. The seawater ratio is approximately 0.320 (Walker et al., 1989); the exact ratio is not important because sample ratios are measured relative to it. Seawater chloride is arbitrarily assigned an isotopic shift value of $\delta^{37}Cl = 0$ per mil (Kaufmann et al., 1984). Significant isotopic variations have been reported for $^{37}Cl/^{35}Cl$ in groundwaters, brines, evaporitic minerals, and fluid inclusions in hydrothermal minerals, with a maximum range in variation in $\delta^{37}Cl$ from -1.3 to +2.2 per mil (Desaulniers et al., 1986; Kaufmann et al., 1984, 1988; Eastoe et al., 1989). At Yucca Mountain, the $^{37}Cl/^{35}Cl$ studies may show that chlorine of meteoric origin has a ratio close to or the same as that of seawater chlorine whereas chlorine originally present in the tuff has a different ratio. Preliminary results obtained during prototype work for this study suggest that the rock chloride is enriched in the heavy isotope, with a $\delta^{37}Cl$ value ~ +1 per mil.

For the purposes of estimating water movement rates and paths, the ^{36}Cl content associated with chlorine of meteoric origin is the only quantity of interest. The ^{36}Cl data from soil profiles will be used to estimate the range of present-day infiltration rates at Yucca Mountain, based upon the extent to which bomb-pulse ^{36}Cl has moved into the soil. The ^{36}Cl data from drillholes and the ESF will be used to establish limits on water movement rates in the deeper subsurface based upon two approaches. First, the uncorrected $^{36}Cl/Cl$ ratio will be used to estimate the upper limit for the water residence time, or the lower limit for the rate of water movement, i.e. the rate of water movement could be faster, but not slower, than indicated by the uncorrected $^{36}Cl/Cl$ ratio. Second, the Cl/Br and/or stable Cl isotopic ratio will be used to estimate the proportion of the chloride that was derived from the atmosphere, as opposed to the rock; and the rock component will be subtracted from the measured ratio in order to estimate the corrected meteoric ratio for the sample. This corrected ratio will then be used to estimate the most probable rate of water movement. The methods to be used for data interpretation are expanded upon in section 3.4.

The use of a bromide salt as a tracer for drilling fluid may pose an additional source of interference for this test. It has been proposed to tag water with about 20 ppm LiBr for dust control of surface and ESF activities. This water would then have a Cl/Br ratio on the order of 1. If salts from the tagged water were to contaminate any of the samples for this test, they would lead one to underestimate the extent of dilution of the meteoric chloride by rock chloride, and hence to overestimate the residence time of water associated with the meteoric chloride (see section 3.4.2 for equations).

3.0 DESCRIPTION OF WATER MOVEMENT TEST STUDY

3.1 General Approach

The key parameter to be measured in this study is the ratio of ^{36}Cl to total chloride as a function of depth in the soluble chloride fraction of soils and rocks and in waters. Ancillary measurements to aid in the interpretation of the $^{36}Cl/Cl$ data include Cl/Br and $^{37}Cl/^{35}Cl$ ratios for these samples, and moisture content and bulk density for the soil samples. The rock and water samples are to be collected from dry-drilled boreholes and from the Exploratory Studies Facility (ESF) at Yucca Mountain. Soil samples are to be collected primarily within the controlled area for the design repository.

3.2 Sample Collection

3.2.1 Soil samples

Two types of soil samples will be collected. First, a pilot suite of about 100 samples will be collected from the surface at Yucca Mountain in order to estimate the degree of variability of the meteoric Cl/Br ratios. Up to 10 of these will also be analyzed for the stable $^{37}Cl/^{35}Cl$ ratio. The results of this pilot study will then be used together with the specified tolerable margin of error at the 95% level of confidence in order to assess the additional number of samples necessary to obtain an estimate within the stipulated margin of error. The Cl/Br and $^{37}Cl/^{35}Cl$ data will aid in interpreting the ^{36}Cl data by providing a geochemical indicator for meteoric chloride that will permit this component to be distinguishable from the indicators for rock chloride. The samples will be collected along several transects within the controlled area as defined in 40 CFR 191.12(g)(1):

> "A surface location, to be identified by passive institutional controls, that encompasses no more than 100 square kilometers and extends horizontally no more than five kilometers in any direction from the outer boundary of the original location of the radioactive wastes in a disposal system..."

The greatest density of sampling will be in the area enclosed by the design repository boundary.

The second type of soil sample to be collected is soil (i.e., alluvial) profiles extending to depths of two or more meters. The purposes of these samples will be: (a) to establish the background meteoric $^{36}Cl/Cl$ ratio, which requires the collection of samples below the ^{36}Cl peak caused by atmospheric testing of nuclear weapons, (b) to test for possible changes in the meteoric Cl/Br and $^{37}Cl/^{35}Cl$ ratios during the infiltration process, and (c) to estimate the infiltration rate at each sample site based on the depth to which the peak of the ^{36}Cl bomb pulse has moved. In two soil profiles studied at Yucca Mountain by Norris et al. (1987), peak values of $^{36}Cl/Cl$ occurred at depths of 0.5 to 1 m; $^{36}Cl/Cl$ ratios returned to background at depths of 1.7 to 2 meters or below. Consequently, profiles will need to be at least two meters deep, and preferably more, to achieve objectives (a) and (c).

One of the criteria for selection of soil sampling sites is a lack of disturbance by man's activities. Where possible, sampling will be done in an opportunistic manner in conjunction with surface-disturbing activities of other SCP field work such as during site preparation for artificial infiltration studies or during drilling of shallow boreholes for neutron moisture meters; such coordination of sampling strategies will facilitate sharing of data and integration of results obtained from related studies. In some cases, a trench will be excavated by a backhoe or bulldozer so as to permit sidewall sampling by hand. In other cases, a truck-mounted auger or other dry-drilling technique will be used. To address objective (c), profile sites will be selected so as to permit the estimation of the average infiltration rate in areas with deep alluvial cover, and an assessment of the spatial variability in this average rate within the controlled area.

The samples collected in this phase of the work will consist of a few kilograms each of soil or alluvium. These will be transported to the Sample Management Facility (SMF) by the user for interim storage and for eventual packaging and shipment to an offsite laboratory for analysis. Precautions will be taken at all steps to protect sample integrity and traceability, in accordance with applicable YMPO and LANL/YMP quality assurance procedures.

3.2.2 Surface-based drillholes

The YMP is planning to drill several boreholes by dry-drilling techniques developed and tested during prototype drilling tests in USW UZ-1 and USW UZ-6 (Whitfield, 1985), at the Apache Leap site in Arizona, and at the Toole site in Utah. Locations of holes planned as of May 1992 are shown in Figure 4; samples from various depths in these boreholes will be requested for ^{36}Cl analyses. Samples will be collected primarily from holes drilled in three drilling programs:

- the systematic drilling program (SD-holes in Figure 4), one objective of which is to provide areal coverage of hydrologic variables in the unsaturated zone;
- the features-based drilling program (UZ-holes in Figure 4), which focuses on evaluating variations in hydrologic properties of the unsaturated zone, including

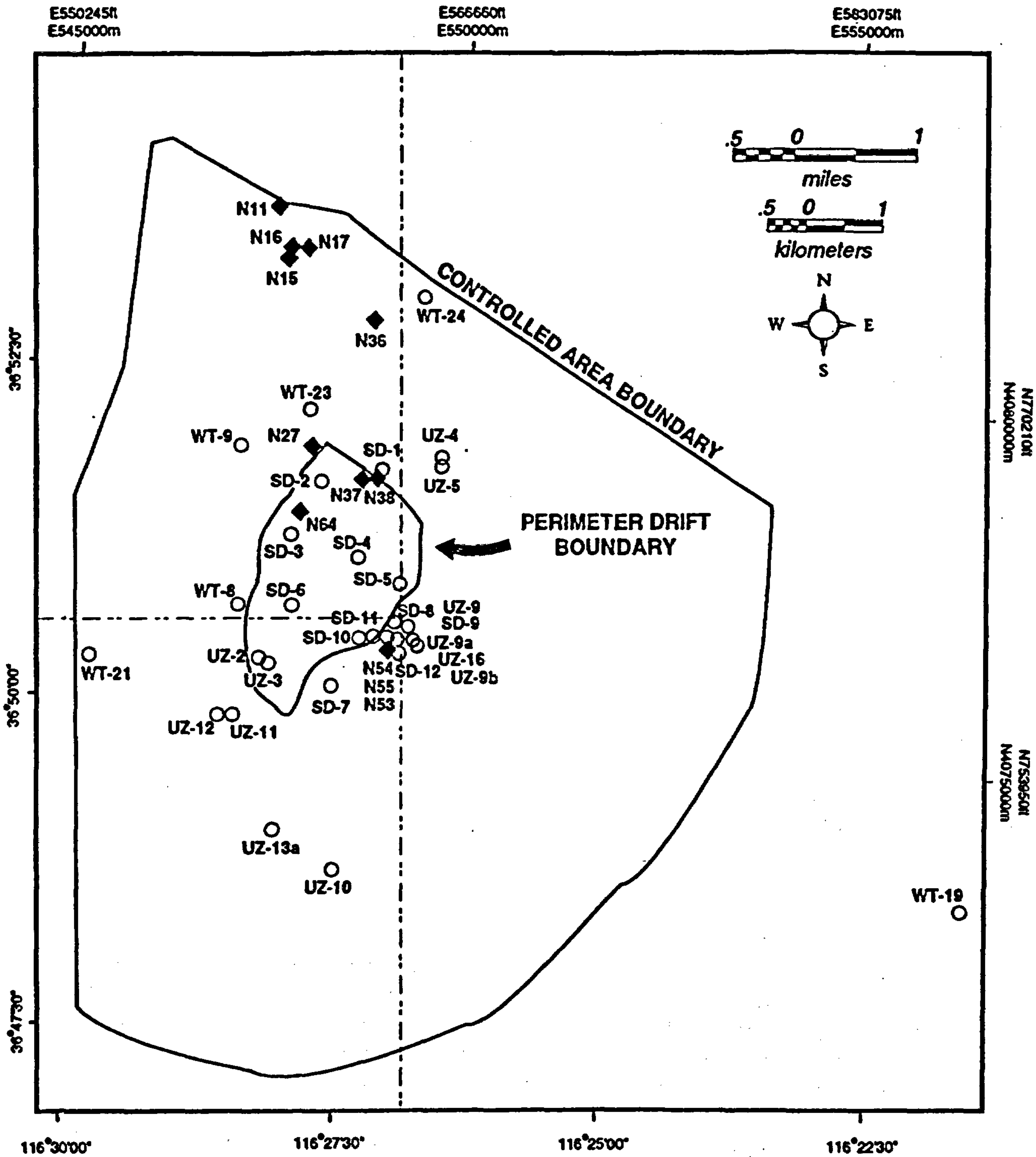

Figure 4. Dry-drilled surface-drilling sites proposed as of May 1992 within the Controlled Area. Neutron-access boreholes shown as solid diamonds; other, generally deeper, boreholes shown as open circles. Map excludes about 12 neutron-access boreholes for which locations were not known as of this date.

the roles of such features as faults, fracture zones and other hydrologic anomalies; and

- the shallow-infiltration drilling program (N-holes in Figure 4), the objective of which is to characterize the statistical distribution of infiltration rates in the uppermost 20-70 m.

Within a given borehole, the sampling strategy is as follows:

- collect at least one sample from each stratigraphic unit in the unsaturated zone, with a maximum interval of 20 m (66 ft) between sample locations;
- collect samples from fault zones;
- collect samples from major fracture zones;
- collect samples from wet zones and perched-water zones;
- collect samples from contacts which are postulated to act as permeability or capillary barriers to downward movement of groundwater; and
- coordinate sampling intervals with other study plan activities as feasible and appropriate, e.g., by collecting cuttings samples from intervals corresponding to core samples to be analyzed for Study Plan 8.3.1.2.2.1, Characterization of Unsaturated-Zone Infiltration, and Study Plan 8.3.1.2.2.7, Hydrochemical Characterization of the Unsaturated Zone.

The strategy will be continually updated as more information is collected, and in response to concerns of the performance assessment modelers. Sampling is expected to become less intense, and more focussed, as outstanding issues are either resolved or more narrowly defined. The borehole stratigraphy of USW G-4 is used to illustrate the selection of sampling depths on the basis of stratigraphic features (Figure 5). In this example, where there are no stratigraphic features to guide the selection of sampling intervals, the maximum interval between samples has been set arbitrarily at 20 m (66 ft).

The quantity of ream-bit cuttings requested from each depth will be specified in documentation prepared by the principal investigator following DOE administrative procedures; the amount will be on the order of 25 kg or more. The field operations branch of the SMF will collect these samples. If at all possible, the samples will not be collected in the form of rock flour, which breaks open saline fluid inclusions and thereby releases rock chloride which dilutes the meteoric chloride during sample preparation. The ideal sample form is as discrete rock chips or core. As shown by the statistical analysis in section 3.6, uncertainties in the age estimates derived by the ^{36}Cl method increase dramatically according to the extent that the sample contains rock chloride. The precision of the method is greatest when the proportion of rock chloride in the sample is minimized.

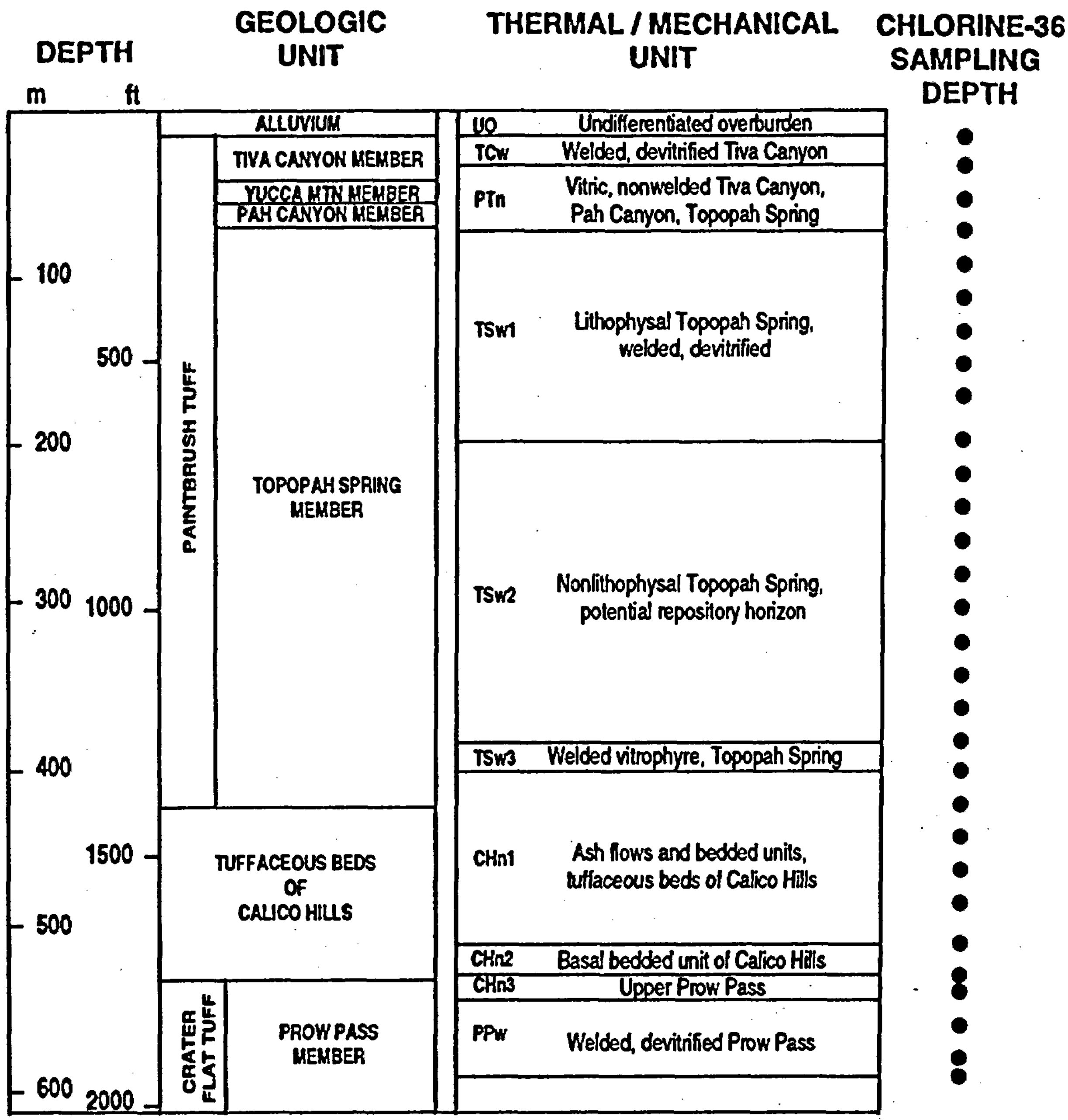

Figure 5. Approximate chlorine-36 sampling depths, shown using the USW G-4 stratigraphic column (based on Ortiz et al., 1985) as an example. Cuttings samples to be collected only above the water table.

3.2.3 Exploratory Studies Facility and mining drifts

Sample collection will also be performed as the Exploratory Studies Facility (ESF) is mined. The tentative test procedure consists of the collection of tuff samples: (a) every 100 m along drifts and ramps, (b) every 30 m along vertical shafts, and (c) as dry-cored samples from contacts, faults and fracture zones. If feasible, for some of the cored samples, $^{36}Cl/Cl$ ratios will be determined as a function of perpendicular distance from the contact, fault or fracture in order to evaluate the role of the geologic feature on water movement. For the cored samples, geologic mapping is required prior to sample collection. Detailed procedures for collection and analysis of all types of samples will be developed prior to the start of ESF construction. The present ESF design is shown in Figure 6 and consists of approximately 12,000 m of mined openings in the Topopah Spring welded unit (the repository horizon) and another 8,000 m in the Calico Hills unit. Thus, on the order of 200 - 300 samples are projected to be collected in the ESF for this study, although not all of these will be analyzed.

The quantity of rock requested from each sampling location will be specified in documentation prepared by the principal investigator following DOE administrative procedures; the amount will be on the order of 25 kg or more. The field operations branch of the SMF will collect these samples. Sample collection methods will also be defined in this documentation and will depend upon the mining method in use:

- If tunnel boring is conducted dry, then the samples for this study will be collected by one of three options: as chips generated by the tunnel boring machine; as core or cuttings from dry-drilling into the tunnel walls; or as rubble from drill and blasting into the tunnel walls.
- If tunneling is conducted with fluid, then all samples for this study need to be collected by dry-drilling into the tunnel walls.
- If mining is by drill and blast methods, then after each designated round, a geologist will select rubble pieces from the working face that are larger than some specified threshold (e.g., 5 cm or 2 in) for transport to the surface before the customary washdown.

Selected samples will be analyzed for chloride, bromide, chlorine-36, and, in some cases, stable chlorine isotopes. In all cases, the containers will be labeled with the depth and spatial coordinates from which the samples were collected, or with unique identifiers allowing the samples to be traced to their collection point.

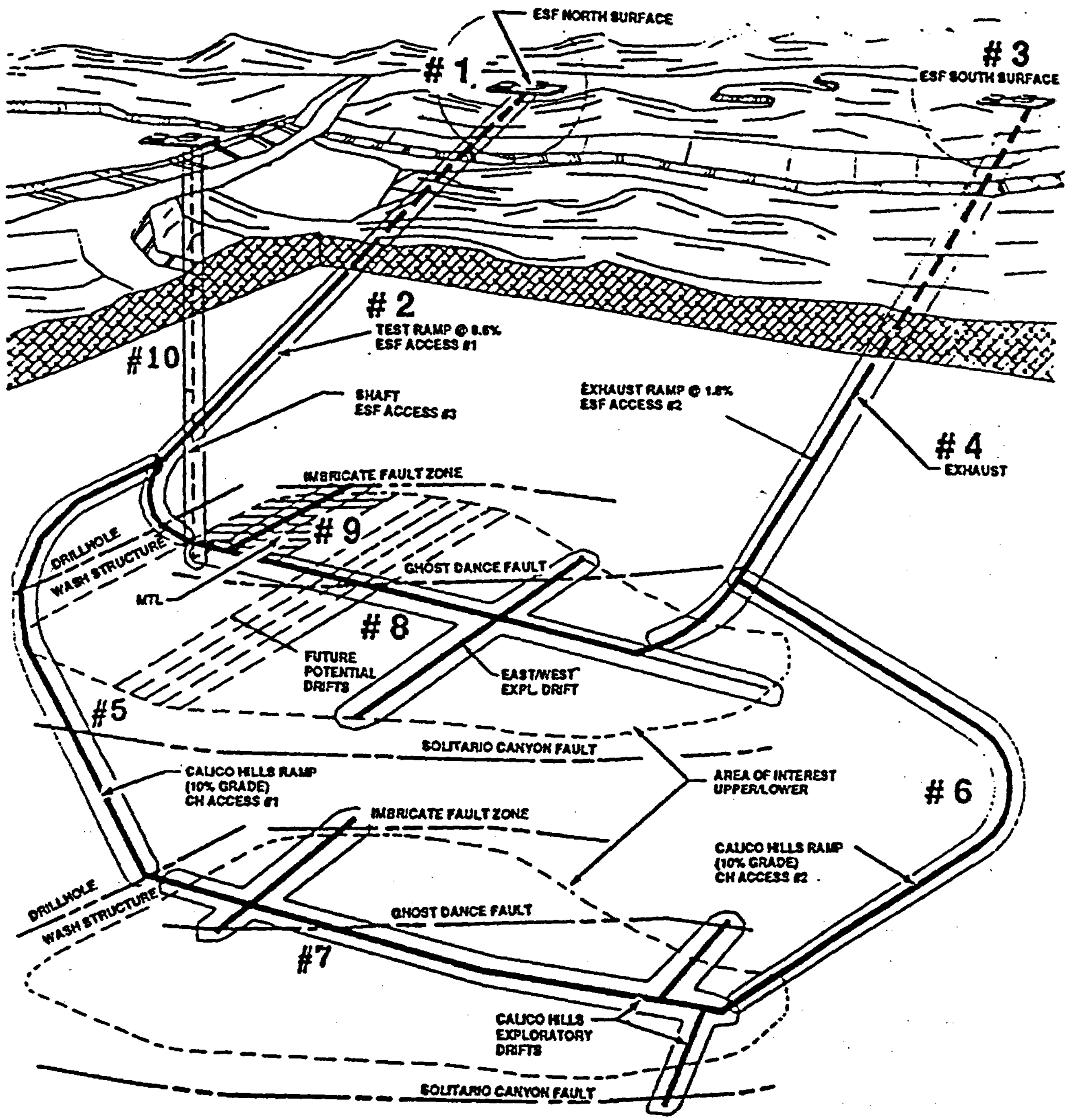

Figure 6. Reference design concept for starting engineering studies for the Exploratory Studies Facility (schematic, not drawn to scale)

3.2.4 Water samples

Water will be collected as available from several different sources: perched water encountered during dry-drilling and mining operations, the water table and deeper zones in the aquifer, and pore water extracted from tuff cores. The ^{36}Cl sampling is dependent upon and must be closely coordinated with the appropriate principal investigators responsible for tritium and ^{14}C age-dating in the unsaturated and saturated zones, as described in the following subsections. Where sufficient sample is available, these water samples will be analyzed for Cl/Br and $^{36}Cl/Cl$, and in some cases also for $^{37}Cl/^{35}Cl$.

3.2.4.1 Perched water

Two types of perched zones are believed to be possible at Yucca Mountain. Near-surface perched zones may develop where water is temporarily impounded in fractures. Such water was observed, for example, during drilling and after completion of some neutron access boreholes on Yucca Mountain. However, this type of perched water is most likely a transient phenomenon and does not add to the net infiltration at Yucca Mountain; the water is imbibed into the matrix and is lost back to the atmosphere by evapotranspiration. A second type of perched water is the possible development of deep perched water zones at lithologic contacts and fault contacts where there are appreciable contrasts in permeability. Larger infiltration fluxes in the past could also result in deep perched water zones. Analyses of Cl/Br and $^{36}Cl/Cl$ in the perched water, when contrasted with the analyses in the matrix overlying and underlying the perched water zone, may be useful in constraining the processes leading to its development and in differentiating between the two types of zones. For a simplistic example, the observation of bomb-pulse ^{36}Cl in a perched water zone with only background levels of ^{36}Cl in the surrounding matrix would indicate that the zone is fed by fracture flow from the surface, as opposed to matrix flow. Other information would of course also influence one's interpretation, such as the lithologic log for the borehole and chemical concentrations in the water. Water samples will be collected from perched water zones under Study Plan YMP-USGS-SP 8.3.1.2.2.3, Characterization of the Yucca Mountain Unsaturated-Zone Percolation: Surface-Based Studies. These water samples are then allocated to various chemical and radiochemical analyses, including $^{36}Cl/Cl$ in this study.

3.2.4.2 Water extracted from cores

Water is being extracted from tuff cores for geochemical and isotopic characterization as part of Study Plan YMP-USGS-SP 8.3.1.2.2.7, Hydrogeochemical Characterization of the Unsaturated Zone. Where the extracted water contains sufficient chloride (e.g., more than 2 mg Cl) following its analysis for other constituents, it will be analyzed for its $^{36}Cl/Cl$ ratio.

3.2.4.3 Water from the saturated zone

Where possible, water will also be collected from below the water table in dry-drilled holes, and from more than one water-bearing interval if these exist. Water samples will be obtained as part of the study plan for Characterization of the Yucca Mountain Saturated Zone Hydrochemistry (YMP-USGS-SP 8.3.1.2.3.2). A single 10-liter sample from each interval is desirable although analyses can be done on smaller quantities; the volume needed is a function of the chloride content and, ideally, the sample should be sufficiently large to contain a minimum of 20 mg chloride to permit replicate analyses for both types of chlorine isotopic analyses. Sampling should occur at least a day or two after cessation of drilling to permit settling of sediments.

3.3 Analytical Methods

In addition to protocols for collection of each type of sample, detailed technical procedures will be followed for the processing of each type of sample to extract soluble halides and to analyze the Cl/Br and chlorine isotopic composition of the extracts. Technical procedures for bulk density and moisture content determinations of soil samples are also needed because values for these parameters are required for interpretation of the soil ^{36}Cl profiles in terms of infiltration rates. Rock moisture contents and bulk densities, when required for this study, will not be measured in this study but rather will be obtained from the YMP technical data base. Soil bulk densities will also be taken from the data base when available; however, soil moisture contents must be analyzed on the same sample used to obtain the ^{36}Cl and Cl/Br analyses and hence will always be determined by this study for the soil profile samples.

The various technical procedures to be used by this study are listed in Table 4; those for sample processing and analysis are summarized in the following sections.

3.3.1 Extraction of halides from soils and rocks

Three methods will be used to extract the soluble halides from soil and rock samples, depending upon the nature of the sample and the desired analyses. Prior to the use of any of the extraction methods, the sample may first need to be sieved to remove large (> 1.25 cm) stones, if experiments confirm that this fraction does not contribute to the soluble halide component. In addition, rock samples that consist of large broken chips will need to be pulverized, such as in a shatterbox, in order to increase the surface area of mineral grains accessible to leaching. However, the extent of pulverization will be minimal in order to minimize breaking open of saline fluid

Table 4. Detailed technical procedures for this study. Note that any of these procedures may be superceded by updated revisions.

PROCEDURE No.	PROCEDURE TITLE	EFFECTIVE DATE
PROCEDURES CONTROLLING SAMPLE COLLECTION (see Note)		
LANL-INC-DP-88	Collection of soil samples for analysis of halides and chlorine-36	01/06/92
PROCEDURES CONTROLLING SAMPLE PROCESSING AND ANALYSIS		
LANL-INC-DP-87	Identification, storage, and handling of samples at Hydro Geo Chem	10/26/92
LANL-INC-DP-89	Procedure for sieving soil and rock samples	01/06/92
LANL-INC-DP-90	Measurement of moisture content of soil samples	06/15/92
LANL-INC-DP-91	Using the shatterbox to pulverize rock samples	In review
LANL-INC-DP-92	Sample leaching to extract soluble chloride and bromide	01/06/92
LANL-INC-DP-93	Step-leaching procedure for extracting soluble chloride and bromide	In review
LANL-INC-DP-94	Using ion chromatography to determine chloride and bromide concentrations	11/30/92
LANL-INC-DP-95	Preparation of samples for chlorine-36 analysis	01/06/92
LANL-INC-DP-96	Measurement of bulk density of soil samples	03/25/92
LANL-INC-DP-97	Preparation of carrier solution for chlorine-36 samples	01/06/92

Note: Water samples will be collected pursuant to study plans 8.3.1.2.2.3, 8.3.1.2.2.7 and 8.3.1.2.3.2. Rock samples for this study will be collected by Sample Management Facility staff.

inclusions which will dilute the meteoric chloride with rock chloride and increase the uncertainty of the water-age estimate (see Section 3.6).

Following sieving and pulverization, three options are available for extracting soluble halides from the soil or rock sample. One is a batch extraction method in which leachable halides are extracted by combining the sample with deionized water, then decanting the solution. Sample size requirements range from as little as 100 g in the case of shallow soil samples intended to be analyzed solely for the Cl/Br ratio, up to 15 or more kg of rock material to be analyzed for chlorine isotopes.

A second method for extracting soluble halides is the column leaching method in which the material is packed into a cylinder through which deionized water flows. This method may be more efficient than the batch extraction technique, producing a smaller volume of leachate that is less turbid.

A third method for extracting soluble halides is the step-leaching procedure in which a sample is leached by batch extraction or column leaching, dried, and then pulverized in a shatterbox. The process is repeated one or more times, with chloride and bromide being measured during each step and chlorine isotopes at less frequent intervals (because of the small yield). The purpose of this procedure is to evaluate the chemical and isotopic characteristics of the rock component of soluble halides. The procedure liberates additional halides with subsequent crushing and leaching such that each step represents a greater component of rock-origin halides and a lesser component of meteoric-origin halides. The data are evaluated by comparing Cl/Br, stable chlorine isotope ratios, or $^{36}Cl/Cl$ ratios versus crushing/leaching step. When the Cl/Br ratios are identical for the last two steps, the values are assumed to represent the pure rock end-member. It is also assumed that the stable chlorine isotope ratios and $^{36}Cl/Cl$ ratios obtained for the latter leaching step represent the rock end-member.

3.3.2 Halide analyses

Chloride and bromide will be assayed in the leachates, in fluids extracted from cores, and in water samples. A wide variety of analytical methods can be used for these two halides. Considerations in the selection of a given method depend upon the detection limit, freedom from interferences, and speed, difficulty, and cost of the analysis. Accuracy and precision are especially critical for this study. High-precision methods applicable to low concentrations of chloride and bromide in water are ion chromatography, colorimetry, and (for bromide) neutron-activation analysis; these and other methods for chloride and bromide are reviewed by Philbert (1982) and Whittemore (1988), respectively. Of these, ion chromatography is the most convenient and is the method of choice for analysis of the field test samples. The procedure requires only a few mL of sample for analysis, requires no sample preparation, and is completed in less than 30 minutes. Its detection limit is 0.01 mg/L or less for chloride and is as low as 1 μg/L for bromide if an anion-concentrator column is used (e.g., Morrow and Minear, 1984).

3.3.3 Chlorine isotope analyses

Processing of the water and leachate samples for analysis of $^{36}Cl/Cl$ and $^{37}Cl/^{35}Cl$ ratios involves the addition of silver nitrate to the samples in order to precipitate silver chloride (AgCl). The only method capable of measuring $^{36}Cl/Cl$ ratios at the levels expected in the Yucca Mountain samples is accelerator mass spectrometry (AMS). Descriptions of the technique are given in Elmore et al. (1984a) and Kubik et al. (1990). For $^{36}Cl/Cl$ ratios on the order of 2×10^{-13} or greater, measurements with precisions of ± 5% are obtained by AMS facilities such as those at the University of Rochester, Lawrence Livermore National Laboratory, and Purdue University. Ratios lower than this value have greater uncertainties, e.g. as shown in Figure 7. The detection limit is 1×10^{-15}. Standards for $^{36}Cl/Cl$ ratios in the range of environmental samples recently became available from the National Institute of Standards and Technology (NIST); development and testing of these standards are described in Sharma et al. (1990). Sample preparation and analysis require careful attention to the possibility for contamination throughout the entire process, from sample collection and storage through sample processing and analysis. A few of the types of contamination problems which have been experienced by other research groups in this field are described in Bird et al. (1990). These problems emphasize the need for quality control procedures, including the frequent preparation of blanks and replicates. The protocol used by the Rochester facility for measuring $^{36}Cl/Cl$ ratios and for estimating uncertainties in the results is described in Elmore et al. (1984b).

Researchers at the Department of Geosciences, University of Arizona, have refined negative ion mass spectrometric techniques for measuring the shift in the stable chlorine isotope ratio, $^{37}Cl/^{35}Cl$, relative to that of seawater, the isotopic standard adopted for such measurements (Eastoe et al., 1989; Kaufmann et al., 1988). Seawater chloride has an isotopic ratio of about 0.320 (Walker et al., 1989). The shift is measured with a precision of ± 0.16 per mil, which is better by a factor of ten or more than previous techniques and is required to detect any variation in the chlorine isotopic composition of most natural samples. The total range of fractionation seen to-date is only about 3 per mil in natural waters (Desaulniers et al., 1986; Kaufmann et al., 1988). The range of fractionation in Yucca Mountain samples is probably somewhat less than this. At one extreme, meteoric chloride should have a ratio identical to that of seawater, i.e., an isotopic shift of 0 per mil. To address the probable ratio for rock chloride, scoping measurements of $^{37}Cl/^{35}Cl$ ratios were made by C. Eastoe (Dept. of Geosciences, University of Arizona, 1990) for samples prepared

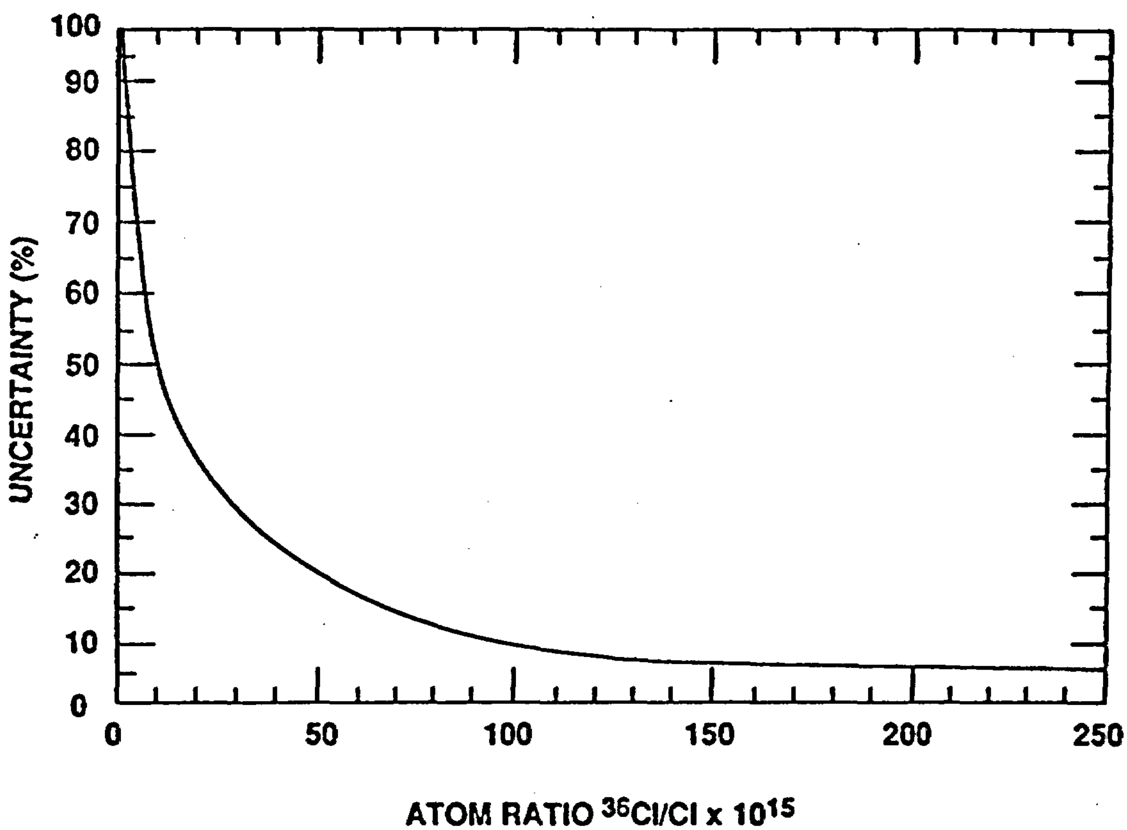

Figure 7. Uncertainties in measured $^{36}Cl/Cl$ ratios as a function of the ratio

from UZ-1 core by Hydro Geo Chem, the YMP subcontractor for this study. The samples judged to contain the greatest proportion of rock chloride had ratios significantly enriched in the heavy isotope, with an isotopic shift of 0.8 to 1.0 per mil. Only one other research group has detected fractionation of chlorine isotopes in natural samples; Vengosh et al. (1989) report a range of fractionation of 8 per mil for samples prepared from brines. However, the precision of their method is only 2 per mil and is thus inadequate for the purposes of this study.

3.3.4 Expected range of measured values

Table 3 (see section 2.2.2) summarizes the parameters that will be measured in this study and their expected range of values. The range of expected values for the $^{36}Cl/Cl$ ratio is determined at the upper end by the contemporary quantity of cosmogenic and thermonuclear ^{36}Cl and at the lower end by the sensitivity of the AMS technique. The ranges of chloride and bromide concentrations are a function of the degree of dilution of the leachate during the extraction of soluble halides from soil and rock samples, and values in Table 3 are based primarily on results from work done to-date by the subcontractor, Hydro Geo Chem, during the development of technical procedures to be used in this study. Any of these ratios ($^{36}Cl/Cl$, Cl/Br, $^{37}Cl/^{35}Cl$) may be shifted outside of the projected range if a source of contamination is present. The expected ranges of soil moisture contents and bulk densities are based on estimates presented in the study plan YMP-USGS-SP 8.3.1.2.2.1, Characterization of the Unsaturated-Zone Infiltration.

3.4 Interpretation of Data

The final part of this test is the analysis and interpretation of the data, which will be conducted as described in the remainder of section 3.4. Details of the models selected for interpretation of the data are expected to change: (a) as data from this study or from studies conducted by other investigators shed light on the validity of the underlying assumptions for a given model; and (b) as the study progresses and better models are developed.

3.4.1 Soil profiles

The data collected from soil samples will serve two purposes: to characterize Cl/Br, $^{37}Cl/^{35}Cl$, and $^{36}Cl/Cl$ ratios in meteoric water, and to provide estimates of present-day infiltration rates. Although the number and location of soil profiles planned to be measured in this study will not be sufficient to estimate the total range of infiltration rates, data from this study will nonetheless expand the data base of such rates applicable to Yucca Mountain.

Infiltration estimates can be derived from data collected in this study by several approaches:

(a) Depth of bomb-pulse ^{36}Cl peak. Average infiltration rates, defined here as net downward linear velocities, can be calculated from the depth of the peak in the bomb-pulse ^{36}Cl concentration profile and the time elapsed from the midpoint in the period of above-ground testing (approximately 35 years). Norris et al. (1987) used this approach to analyze data from a soil profile in Yucca Wash and calculated a linear velocity of 18 mm/yr; assuming a volumetric water content of 10%, this value corresponds to a net infiltration rate of 1.8 mm/yr.

(b) Depth of center of mass of bomb-pulse ^{36}Cl. Where moisture contents or bulk densities are measured, more precise estimates of water velocities can be calculated from the depth of the center of mass of the bomb-pulse ^{36}Cl concentration profile, z_0, as described by Walker et al. (1991), whose mathematical development is presented here. The center of mass is defined by

$$\int_0^{z_0} \theta(z,t)\ c(z,t)\ dz = \frac{M}{2} \qquad (1)$$

where

$\theta(z,t)$ = volumetric water content at depth z and time t (dimensionless),
$c(z,t)$ = concentration of the tracer in the soil water (g cm^{-3}), and
M = total solute mass (g cm^{-2}) (i.e., the integral of bomb-pulse ^{36}Cl) in the soil.

Taking the total derivative of this equation with respect to time and assuming conservation of mass (i.e., dM/dt = 0), one obtains

$$\frac{dz_0}{dt} = \frac{q_s(z_0,t)}{\theta(z_0,t)\ c(z_0,t)} = \frac{q_w(z_0,t)}{\theta(z_0,t)} = \overline{V}(z_0,t) \qquad (2)$$

where

$q_s(z_0,t)$ = solute flux at depth z_0 and time t (g cm^{-2} s^{-1}),
$q_w(z_0,t)$ = water flux at depth z_0 and time t (cm^3 cm^{-2} s^{-1}), and
$\overline{V}(z_0,t)$ = mean pore water velocity (cm s^{-1}).

An underlying assumption is that the tracer movement mirrors the water movement, which is valid provided the average soil water concentration of the tracer at z_0 equals that of the water below z_0. This approximation is affected by such processes as anion exclusion, retardation, diffusion, aggregate dispersion and vapor movement of water

(Walker et al., 1991) and is invalid if bypass flow or preferential pathways dominate transport through this zone (Scanlon, 1992b; see also Section 3.4.3).

(c) If the bomb-pulse ^{36}Cl profile extends well below the root zone, then the above approach may provide an estimate of the recharge rate, which is expected to be some small fraction of the infiltration rate.

(d) Chloride mass balance accumulation time. The stable chloride concentration profile can also be used to estimate water transport rates. This approach has been widely used, e.g. Sharma and Hughes (1985), Matthias et al. (1986), Phillips et al. (1988), Scanlon et al. (1990) and Scanlon (1991). The travel time of chloride in the profile, assumed to represent the travel time of water, is evaluated according to

$$t = \frac{\Sigma_i \, (Cl_i \, z_i)}{P \, Cl_p} \qquad (3)$$

where Σ_i (Cl_i z_i) is the cumulative total mass of chloride from the surface to depth z_i, P is the mean annual precipitation rate, and Cl_p is the mean chloride concentration in precipitation, including the contribution from dry fallout. The latter variable may be estimated by dividing the natural ^{36}Cl fallout at the site by the product of mean annual precipitation and the natural (prebomb) $^{36}Cl/Cl$ ratio for that site (Phillips et al., 1988; Scanlon et al., 1990). The approach accounts for loss of water by evapotranspiration but assumes that chloride acts conservatively. For the vicinity of Yucca Mountain, mean annual precipitation is 15 cm/yr, the prebomb ratio is about 5.2×10^{-13} (Norris et al., 1987), and the natural ^{36}Cl fallout is estimated by Bentley et al. (1986a) to be about 28 atoms ^{36}Cl m^{-2} s^{-1}, resulting in an estimated Cl accumulation rate (P Cl_p) of 100 mg m^{-2} yr^{-1}, with a corresponding concentration in precipitation of 0.67 mg/L for Cl_p. These estimates will be revised as more data become available from this and other Yucca Mountain studies.

(e) Qualitative information may also be provided by the measured soil profiles. Together with the degree of smoothness of the profile, the integral of the bomb-pulse ^{36}Cl concentration peak can be compared against the expected integral for total fallout at the site to provide an indication of the variability of rainfall or evidence for preferred infiltration pathways (e.g., Norris et al., 1987).

3.4.2 Rock samples from boreholes and the Exploratory Studies Facility

As in the case of the soil profiles, ^{36}Cl data from rock samples can be interpreted by several approaches.

(a) The Limiting Model. In the simplest case, the measured $^{36}Cl/Cl$ ratios can be used to estimate upper limits for average water residence time at the sampled depths, and hence lower limits for the net downward velocities or fluxes to those depths. The linear downward velocity is defined as vertical depth divided by ^{36}Cl age and thus implies the assumption that water moves vertically through the matrix. The linear velocity can be used to estimate the Darcy velocity or Darcy flux by multiplying the velocity by the volumetric water content of the intervening rock units; this conversion of results may not be straightforward where the water contents vary considerably from one unit to another.

For the ^{36}Cl-based age estimates, the assumptions are that the initial meteoric $^{36}Cl/Cl$ ratio is known and constant during the period of interest. For old water, the buildup of ^{36}Cl as a result of the subsurface neutron flux must also be taken into account. The secular equilibrium $^{36}Cl/Cl$ ratio is on the order of 2×10^{-14} and can be determined most precisely by measuring the ratio in rocks themselves, after first removing any meteoric component of ^{36}Cl by leaching. Then an age estimate is obtained by applying the standard equation for radioactive decay with correction for ingrowth (Bentley et al., 1986a)

$$t = - \frac{1}{\lambda_{36}} \ln \left[\frac{(R_x)_{36} - (R_r)_{36}}{(R_0)_{36} - (R_r)_{36}} \right] \tag{4}$$

where λ_{36} is the decay constant for ^{36}Cl, 2.30×10^{-6} yr^{-1} (Walker et al., 1989), and $(R_x)_{36}$, $(R_r)_{36}$ and $(R_0)_{36}$ are the measured, rock, and initial $^{36}Cl/Cl$ ratios, respectively.

Data from Norris et al. (1990) can be used to illustrate this approach. The lowest measured $^{36}Cl/Cl$ ratio in borehole UZ-1 was 1.0×10^{-13} at a depth of 372 m. For an initial recharge $^{36}Cl/Cl$ ratio of 5.2×10^{-13} and a rock $^{36}Cl/Cl$ ratio of 2×10^{-14}, this value sets an upper limit for average residence time of 8.0×10^{5} yr at this depth and a minimum net downward linear velocity of 0.5 mm/yr. For 10% moisture content in the Topopah Spring unit, this value corresponds to a lower limit for the net downward flux of 0.05 mm/yr.

(b) Dilution Model. A more realistic estimate of residence time and net velocity can be obtained by correcting the measured $^{36}Cl/Cl$ ratio for dilution of the meteoric component by rock chloride, which is introduced into the sample as a consequence of drilling and crushing in the laboratory. Rock chloride generally has a $^{36}Cl/Cl$ value that is more than an order of magnitude lower than that of meteoric chloride. It is hypothesized that the proportion of rock chloride may be estimated by assuming two-component mixing between meteoric and rock chloride. Each of these end-members is assumed to have a characteristic and constant Cl/Br and/or $^{37}Cl/^{35}Cl$ ratio. This assumption will be tested as soil and rock samples become available from Yucca Mountain. In addition, for the purposes of calculations presented here, the rock component is assumed to have a characteristic and constant $^{36}Cl/Cl$ ratio, an assumption which will also be continually re-evaluated as rock samples come available. Any systematic variations in these end-member ratios, e.g., due to elemental variations as a function of location or rock unit, may necessitate a more refined treatment of the data (see section 2.2.5.2).

According to this dilution model, the measured Cl/Br, $^{37}Cl/^{35}Cl$ and $^{36}Cl/Cl$ ratios, $(R_x)_i$, are determined from the generic mixing equation

$$(R_x)_i = f_m\,(R_m)_i + f_r\,(R_r)_i \qquad (5)$$

where f_m and f_r are the relative proportions of chloride derived from meteoric and rock sources, respectively, and are assumed to sum to 1; and R_m and R_r are the end-member ratios for meteoric and rock chloride, respectively. The subscript i refers to the type of ratio: $^{36}Cl/Cl$ for i = 36, CB for i = Cl/Br, or $^{37}Cl/^{35}Cl$ for i = 37/35. The value of f_m and hence f_r can be estimated from the measured Cl/Br and/or $^{37}Cl/^{35}Cl$ ratios by solving the mixing equation for f_m. Because the uncertainties in the $^{37}Cl/^{35}Cl$ measurements are fairly large (± 0.16 per mil) relative to the total range of variation expected in the Yucca Mountain samples (about 1 per mil, see table below), f_m will probably be estimated from the Cl/Br ratios,

$$f_m = \frac{(R_x)_{CB} - (R_r)_{CB}}{(R_m)_{CB} - (R_r)_{CB}} \qquad (6)$$

Measured $^{37}Cl/^{35}Cl$ ratios may be used as confirmatory data for selected samples. Once a value of f_m has been estimated for a given sample, the meteoric $^{36}Cl/Cl$ ratio, $(R_m)_{36}$, can be estimated from the mixing equation,

$$(R_m)_{36} = \frac{(R_x)_{36} - f_r\,(R_r)_{36}}{f_m} \qquad (7)$$

To-date, the best available estimates for the end-member values of the various ratios are as shown in Table 5, and will be continually revised as more data comes available from this study. Statistical analysis of variance and covariance may also be applied in order to assess whether any of these end-member values vary systematically, e.g., as a function of location, stratigraphic unit, or degree of hydrothermal alteration.

Table 5. Estimated end-member values for Cl/Br and chlorine isotopic ratios in halides derived from meteoric and rock sources at Yucca Mountain

Tracer	Meteoric end-member, R_m	Rock end-member, R_r	Estimated uncertainties in end-member values
Cl/Br	130 (Norris et al., 1990)	500 (Norris et al., 1990)	± 5%
$^{37}Cl/^{35}Cl$	0.0 per mil	1.0 per mil	Unknown
$^{36}Cl/Cl$	5×10^{-13} for prebomb chloride (Norris et al., 1990)	2×10^{-14} (Norris et al., 1990)	± 20%

NOTE: Estimates of end-member values and uncertainties will continually be revised as additional data are collected in this study.

To apply the dilution model, the Cl/Br and/or $^{37}Cl/^{35}Cl$ ratio must be measured in the same rock leachate as is used to measure the $^{36}Cl/Cl$ ratio. Unfortunately, such paired measurements are not available for the analyses reported in Norris et al. (1990) for borehole UZ-1. However, a few paired analyses have been made by Hydro Geo Chem, the subcontractor for this study. The significance of this correction procedure can be illustrated using one of these pairs, obtained for the sample from the depth of 77 m in UZ-1. The uncorrected $^{36}Cl/Cl$ ratio $(R_x)_{36}$ is 1.9×10^{-13} for the second step in the step-leaching procedure (unfortunately, this ratio was not measured in chloride extracted during the first step). Using equation (4), this ratio corresponds to a maximum residence time of 4.7×10^5 yr and a minimum net downward linear velocity of 0.16 mm/yr. The measured Cl/Br ratio for this leachate fraction is 269, suggesting that 62% of the chloride leached from this rock sample was meteoric in origin, i.e., $f_m = 0.62$ by equation (6). The corrected meteoric $^{36}Cl/Cl$ ratio $(R_m)_{36}$ is then 2.9×10^{-13} from equation (7). Substituting $(R_m)_{36}$ for $(R_x)_{36}$ in equation (4), one obtains an estimated residence time of 2.7×10^5 yr and an average downward linear velocity of 0.29 mm/yr, nearly twice the lower limit established by the previous method. This approach thus provides best estimates, as opposed to limits, for the flux. (Note that the cited example was not obtained using approved technical procedures or following quality assurance procedures of the caliber required by the YMP.)

(c) Qualitative information may also be provided by the measured profiles. The observance of bomb-pulse $^{36}Cl/Cl$ ratios at depth might provide evidence for the role of specific faults, lineaments or fractures as routes for rapid infiltration of water or, alternatively, as barriers to flow. Fracture flow and lateral flow are particularly expected through the Tiva Canyon and Pah Canyon Members. If ^{36}Cl values higher than background are encountered in samples from surface-based boreholes and from the Exploratory Studies Facility, the data will be examined to determine if fracture flow or lateral flow might account for the inclusion of bomb-pulse ^{36}Cl. An irregular profile of $^{36}Cl/Cl$ with depth in a given formation could indicate that flow paths are strongly influenced by the fracture system. The distribution of flux estimates in the deep subsurface may also provide a means for evaluating whether paleorecharge rates were greater in the past than at present, which might be expected since precipitation rates were greater. In all cases, the ^{36}Cl data must be examined in concert with other independent lines of evidence—such as lithologic logs, hydraulic parameters and other isotopic and geochemical data—in order to ensure that the interpretation is consistent with all available information. Wherever possible, for example, ^{36}Cl sampling will be coordinated with that for ^{14}C and tritium. However, discordant ages from different isotopic techniques is to be expected as a consequence of the inevitable mixing of waters of different ages (e.g., Davis and Murphy, 1990; Fröhlich et al., 1991), an effect that may be enhanced at Yucca Mountain due to the presence of preferential pathways in the flow system as well as by differences in geochemical behaviors of the various tracers.

(d) Variance-component estimation and trend estimation. Where justified by the data, analysis of variance will be performed in order to quantitatively assess the dominant controlling variables for water residence times and, by extension, for water flow paths and rates. Some of the conceptual flow hypotheses listed in Table 2 will be tested by assessing whether differences between ^{36}Cl-based estimates of residence times are statistically significant:

- for samples from fractures and fracture zones relative to those from adjacent unfractured matrix;
- for samples from faults relative to those from adjacent matrix;
- for samples from opposite sides of a fault, in order to assess role of fault as possible barrier to flow;
- for samples from perched water zones relative to those from the matrix immediately above these zones; and
- for samples immediately above and below lithologic contacts.

Other statistical tests will be applied to evaluate the presence of lateral flow, such as along lithologic contacts, by testing for spatial trends in residence times of samples from the same stratigraphic depth. As work progresses, other variables may also be identified and tested for significance in controlling the spatial variability of residence time. To the extent that the data warrant more detailed treatment, multivariate regression methods may be used to develop a predictive model for identifying trends. Absolute and relative water residence times determined from this study for different zones of Yucca Mountain will be compared to those predicted by transport models.

3.4.3 Limitations and assumptions for the interpretation of the ^{36}Cl data

The interpretation of data as described in the previous section must recognize the limitations of the technique as well as the simplifying assumptions implicit in the models. These limitations and assumptions are discussed below. The validity of these assumptions will be re-evaluated as additional data come available. Where an assumption is found to be invalid or questionable, more refined models may need to be developed to interpret the ^{36}Cl and chloride data.

3.4.3.1 Effective dating range

The $^{36}Cl/Cl$ analyses can provide useful information for unsaturated zone studies if an unambiguous bomb-pulse signal is detected, or if an unambiguous decay of the meteoric signal can be shown. The method is insensitive if the hydrologic response time is greater than forty or less than about 100,000 years. Thus, for example, if the water residence time in the deep subsurface (beyond the depth of the bomb-pulse signal) is much less than 100,000 years, then ^{36}Cl-based tests of the flow hypotheses listed in Table 2 and discussed above may not be useful.

3.4.3.2 Flow field

Models for estimating infiltration rates based upon the depth of bomb-pulse ^{36}Cl or upon the accumulation of chloride in soil profiles assume one-dimensional, vertically downward, piston-type flow at steady state (e.g., Johnston, 1987; Scanlon, 1991, 1992a). At some field sites, soil textures such as the formation of clayey zones or silcrete layers may cause lateral flow, invalidating the assumption of one-dimensional downward movement of water. To a limited extent, more complicated variants of the models may allow for somewhat less stringent requirements, e.g., non-steady-state and two-dimensional flow. Preferential pathways can be formed by plant roots, animal activity, or fissured sediments, and could invalidate the assumption of piston-type flow (e.g., Beven and Germann, 1982; Bowman and Rice, 1986; Johnston, 1987; Roth et al., 1991; Scanlon, 1992b). Knowledge of the nature of the bypass flow may allow bounds to be established on its effect on the infiltration rate (e.g., Roth et al., 1991).

3.4.3.3 Constancy of end-member values

A premise of data interpretation for this study is that the meteoric $^{36}Cl/Cl$ ratio has been constant throughout the Quaternary Period, excluding the enhanced ^{36}Cl fallout following atmospheric testing of nuclear devices between 1952 and 1958. This premise is akin to assuming that ^{36}Cl production rates in the atmosphere and the rate of fallout of stable Cl at the site has been constant during that period. Both of these assumptions are likely to be invalid to some extent; what is not known is the magnitude of the deviations from constancy. The ^{36}Cl production rate is controlled by the flux of galactic protons, solar-wind magnetic properties, and the dipole moment of the earth. The galactic flux is generally assumed constant; changes in the solar-wind modulation are poorly known but have not been unambiguously documented. However, the strength of the Earth's magnetic field is known to have varied during the Holocene up to twice its present value, which would affect the production rate of ^{36}Cl and other cosmogenic nuclides because cosmic-ray ions are deflected by this magnetic field, i.e., the stronger the field, the lower the rate of nuclide production (Bard et al., 1990). The effect on ^{14}C production rate is on the order of 10-20% (Bard et al., 1990). The effect on ^{36}Cl production rate is not known but could be larger than that for ^{14}C because the atmospheric inventory of ^{36}Cl is not buffered against changes by the ocean inventory, as is the case for ^{14}C. Evidence bearing on variations in ^{36}Cl production rates is being sought in natural archives of these records such as polar ice and pack-rat middens (e.g., Elmore et al. 1984a; Phillips et al., 1991). For the present study, fluctuations in $^{36}Cl/Cl$ profiles obtained in thick alluvial material will provide data for estimating the magnitude of changes in the meteoric ratio.

The $^{36}Cl/Cl$ ratio representative of the rock end-member may be considerably higher in near-surface rocks due to in-situ production by cosmogenic processes (Phillips et al., 1986b; Zreda et al., 1991). Cosmogenic production by spallation of calcium and potassium isotopes and by neutron-capture by ^{35}Cl needs to be considered as a correction term for rock samples collected from within one or two meters of the surface.

The model presented in section 3.4.2 also assumes that Cl/Br and/or $^{37}Cl/^{35}Cl$ ratios of the meteoric and rock end-members are constant values. This assumption may not be valid. For example, end-member ratios for the tuffs may vary among the various lithologic units, or as a function of the extent of hydrothermal alteration of a given unit or subunit. This issue may be addressed by analyzing a sufficient number of samples from a variety of locations to conduct an analysis of variance and spatial-trend analysis. Although it is not expected that the present-day meteoric end-member Cl/Br ratio will vary spatially within the controlled area, this ratio may conceivably have been different in the past at Yucca Mountain under different climatic regimes when different storm tracks may have been dominant because the Cl/Br ratio varies as a function of distance from the oceanic source. This issue may be addressed by sampling surface soils for Cl/Br in a broader region surrounding Yucca Mountain and assessing the magnitude of the variability. It is also possible that marine aerosols are not the important determinant of meteoric chloride deposition rates at Yucca Mountain (e.g., Lewis et al., 1984), in which case other factors may govern its accession rate and variability in halide ratios. Such a possibility can be assessed by reviewing deep alluvial profiles of halide concentrations and ratios.

3.4.3.4 Differential transport rates for chloride and water

Interpretation of the ^{36}Cl data in terms of the rate of water movement through the unsaturated zone requires the consideration of processes that may differentiate chloride movement from water movement. Theoretically, the rate of movement of a tracer introduced into the unsaturated zone, even a perfectly conservative tracer, will lag behind that of the water front because of dilution of the tracer with the initial water content. Hence, tracer movement will approximate water movement only if the initial water content is negligible or if changes in moisture content are sufficiently small, i.e., it will lag water movement under sudden recharge events.

Another process which could lead to differential rates of movement for water and chloride 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 water; this effect has been documented for chloride and ^{36}Cl in soils by numerous researchers (James and Rubin, 1986; Krupp et al., 1972; Smith, 1972; and Thomas and Swoboda, 1970) and for ^{36}Cl in saturated alluvium by Ogard et al. (1988). The effect is maximized in unsaturated soils with a high clay content, in which the average anion velocity may be as much as twice that of the water; in unsaturated sand, which is closer in composition to the alluvial cover at Yucca Mountain, the enhancement in transport rate may be about 5-10% and is highest for low water contents (James and Rubin, 1986). The latter authors present a solute transport model which includes anion exclusion effects. However, the effect of anion exclusion may have been over-estimated in the various cited studies because the water velocities in the experiments were generally determined by monitoring the transport rate of tritiated water, but no consideration was taken to evaluate the extent to which tritiated water molecules may be retarded relative to non-tritiated water molecules.

Anion-adsorption or assimilation of chloride by plant roots are other possible factors which would act in the opposite direction, retarding the rate of chloride movement relative to that of water. Anion-adsorption is expected to be negligible in the Yucca Mountain environment because it is generally important only at low pH values (typically below pH 5) while soils at Yucca Mountain are generally in the basic range. The role of plant roots in retarding chloride movement to a greater extent than it does water movement requires additional evaluation by this study.

Another aspect of chloride movement that is important for hydrologic modeling and that is not negligible arises from the nonvolatile character of these anions. Water movement through the unsaturated zone may occur in downward pulses through the matrix in the liquid phase, followed by upward movement in the vapor phase, but chloride ions move only in the liquid phase. Hence, while the net flux might actually be upwards, the ^{36}Cl data can only provide an estimate of the downward component. In addition, the ^{36}Cl decay data will probably reflect an age that results from mixing chloride ions from more than one pulse. Nonetheless, if ^{36}Cl-based travel times for samples from below the repository horizon are significantly greater than those for samples from the repository horizon, then these data may be appropriate as an analog for estimating the average travel time of a radioactive waste nuclide such as ^{99}Tc, which is expected to be transported in water as the TcO_4^- ion. Like chloride, the pertechnetate ion is both nonvolatile and nonsorbing.

Dispersion of bomb-pulse ^{36}Cl was found to be significant in desert soil profiles by Phillips et al. (1988). These authors found that a dispersivity of 8 cm used in their finite-element solute transport model provided the best match to the observed ^{36}Cl distribution with depth. The preliminary interpretation of soil ^{36}Cl data from this study, using the simplistic model described in section 3.4.1(b), will neglect dispersion (e.g., see evaluation by Scanlon, 1991), but a more detailed evaluation of the data following the approach of Phillips et al. (1988) may be warranted.

Some researchers have proposed that plant roots cycle chloride in and out of soil water as necessary to maintain an appropriate osmotic potential in their microenvironment. In this case, chloride may have a residence time in the root zone that is considerably longer than that of water, such that the models described in section 3.4.1 for soil profiles give rise to ages that are larger, and infiltration rates that are lower, than the true values.

3.4.3.5 Interferences from other field activities

Data interpretation is conducted under the assumption that any interference from other field activities, as discussed in section 2.2.5, will be readily recognized.

3.5 Equipment and Services Required

No specialized equipment or services are required to conduct this study. No surface-based drilling or trenching is required to be initiated for this study; all surface-based sampling activities will be conducted using access provided through drilling or trenching conducted as part of other site-characterization activities. Support for sample collection in the ESF will be coordinated through the Test Coordination Office. Preparation and analysis of samples will be conducted by a subcontractor following the technical procedures described in section 3.3 and listed in Table 4.

3.6 Estimated Degree of Accuracy of Results

The $^{36}Cl/Cl$ data, when plotted against sample depth, provide a measure of water velocity down through the unsaturated zone, assuming vertical flow through the rock matrix. The precision of the estimated velocities depends upon the precision with which each parameter can be determined. The sensitivity analysis presented in this section serves to emphasize the importance of high-precision numbers for all of the analyses to be made in this study. The $^{36}Cl/Cl$ ratios of the samples are only one parameter among many; uncertainties in Cl/Br ratios for the sample and end-members may contribute significantly to the total uncertainty of the final result. The sensitivity analysis also underscores the need to minimize leaching of the rock component of Cl from the sample because the uncertainty of the final result increases dramatically in proportion to the extent of dilution of the meteoric Cl by rock Cl.

The following statistical analysis illustrates the method by which uncertainties will be estimated for the borehole and ESF sample results and provides a preliminary indication of the degree of precision expected for the velocity estimates. The estimated magnitude of the uncertainty will be re-calculated periodically as information is obtained to evaluate the limitations and assumptions of the dating method described in section 3.4.3.

Uncertainties in estimating the proportion of meteoric chloride in a sample. The mixing equation (6) is used to estimate the proportion of meteoric Cl, f_m, from the measured end-member and sample Cl/Br ratios, R_m, R_r, and R_x, respectively. The uncertainty in f_m is approximated by taking the square of the total derivative of Equation (6), ignoring the cross terms (Young, 1962, pp. 96-98):

$$\sigma_{f_m} = \left[\left(\frac{\partial f_m}{\partial R_x}\right)^2 \sigma^2_{R_x} + \left(\frac{\partial f_m}{\partial R_m}\right)^2 \sigma^2_{R_m} + \left(\frac{\partial f_m}{\partial R_r}\right)^2 \sigma^2_{R_r} \right]^{0.5} \qquad (8a)$$

$$= \frac{\left[(R_m-R_r)^2\ \sigma^2_{R_x} + (R_x-R_r)^2\ \sigma^2_{R_m} + (R_m-R_x)^2\ \sigma^2_{R_r}\right]}{(R_m - R_r)^2} \qquad (8b)$$

Uncertainties associated with the Cl/Br ratios for the end-members and samples will be estimated following the detailed technical procedure for those analyses. Figure 8 shows how the relative uncertainty in f_m is expected to increase with the analytical uncertainty as well as with the extent of dilution of the meteoric Cl by rock Cl.

Uncertainties in the estimate of the meteoric $^{36}Cl/Cl$ ratio for a given sample. Equation (7) is used to estimate the meteoric $^{36}Cl/Cl$ ratio, R_m, based on the measured $^{36}Cl/Cl$ ratio, R_x, and the estimate of f_m and of the $^{36}Cl/Cl$ ratio of the rock end-member, R_r. Uncertainties associated with the meteoric $^{36}Cl/Cl$ ratio are estimated by taking the square of the total derivative of equation (7), again ignoring cross-terms:

$$\sigma_{R_m} = \left[\left(\frac{\partial R_m}{\partial f_m}\right)^2 \sigma^2_{f_m} + \left(\frac{\partial R_m}{\partial R_x}\right)^2 \sigma^2_{R_x} + \left(\frac{\partial R_m}{\partial R_r}\right)^2 \sigma^2_{R_r}\right]^{0.5} \qquad (9a)$$

$$= \left[\left(\frac{(R_x - R_r)^2}{f_m^{\,2}}\right)^2 \sigma^2_{f_m} + \left(\frac{1}{f_m}\right)^2 \sigma^2_{R_x} + \left(\frac{f_m - 1}{f_m}\right)^2 \sigma^2_{R_r}\right]^{0.5} \qquad (9b)$$

Uncertainties in measured $^{36}Cl/Cl$ ratios for samples are approximated according to the method in Elmore et al. (1984a) and should be on the order of those shown in Figure 7. For purposes of illustration, the $^{36}Cl/Cl$ ratio for the rock end-member is assumed to be 2.0 (± 0.4) x 10^{-14}. Figure 9 shows how the relative error in the meteoric $^{36}Cl/Cl$ ratio is expected to vary as a function of f_m.

Uncertainties in the estimated residence time. The water residence time is estimated based on applying the dating equation (4) to the meteoric $^{36}Cl/Cl$ ratio after substituting R_m for R_x. The following expression is derived by taking the square of the total derivative of equation (4), and ignoring the cross terms:

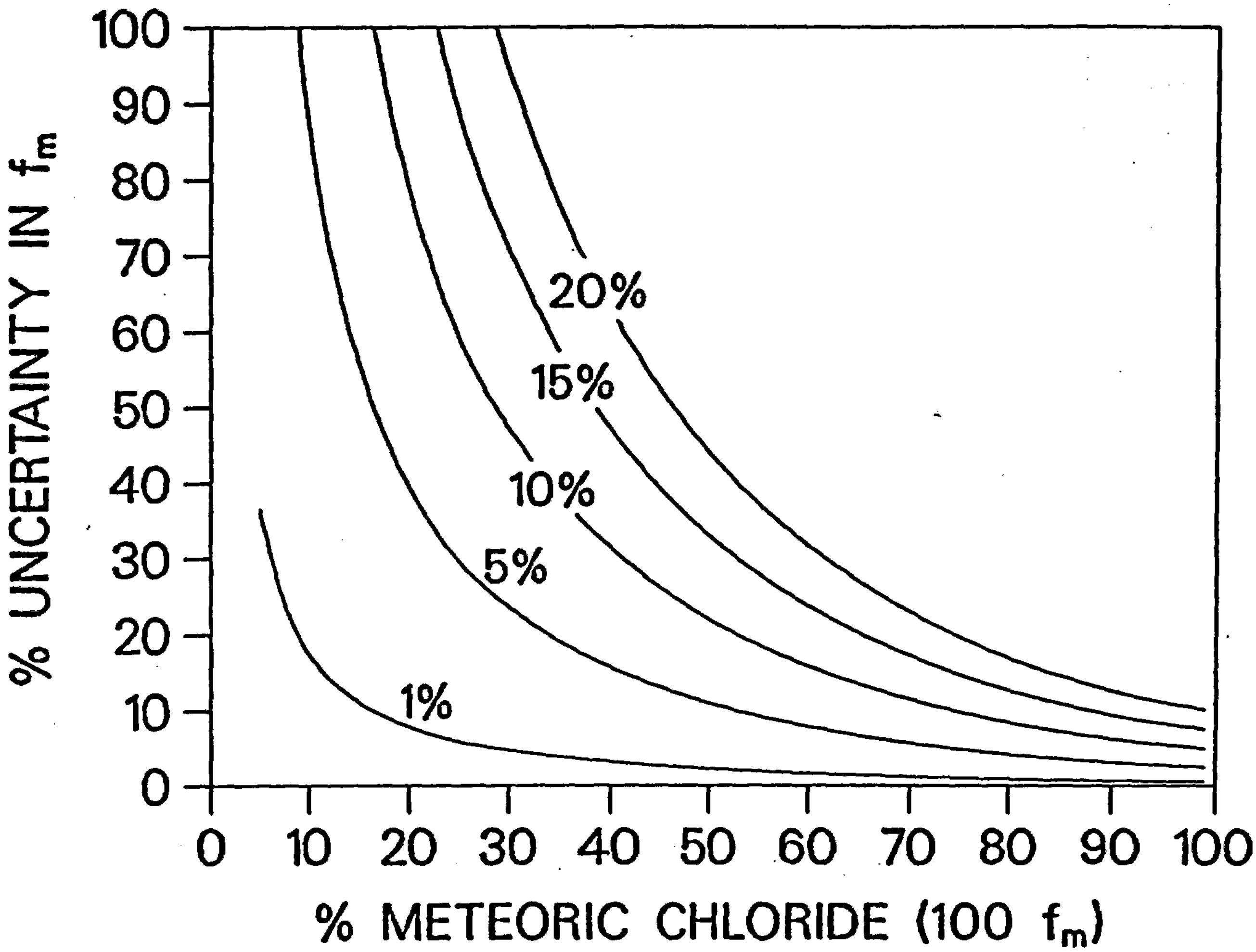

Figure 8. Relative uncertainty in the estimated proportion of meteoric chloride, f_m, in a sample as a function of the analytical precision of the Cl/Br determinations. Curves are labelled with the relative errors assumed for the Cl/Br analyses. The same relative precisions (σ_R/R) is assumed to apply to the meteoric and rock ratio end-members as well as to the sample ratio. Calculated from equations (6) and (8b) assuming Cl/Br ratios of 130 and 500 for the meteoric and rock ratios, respectively.

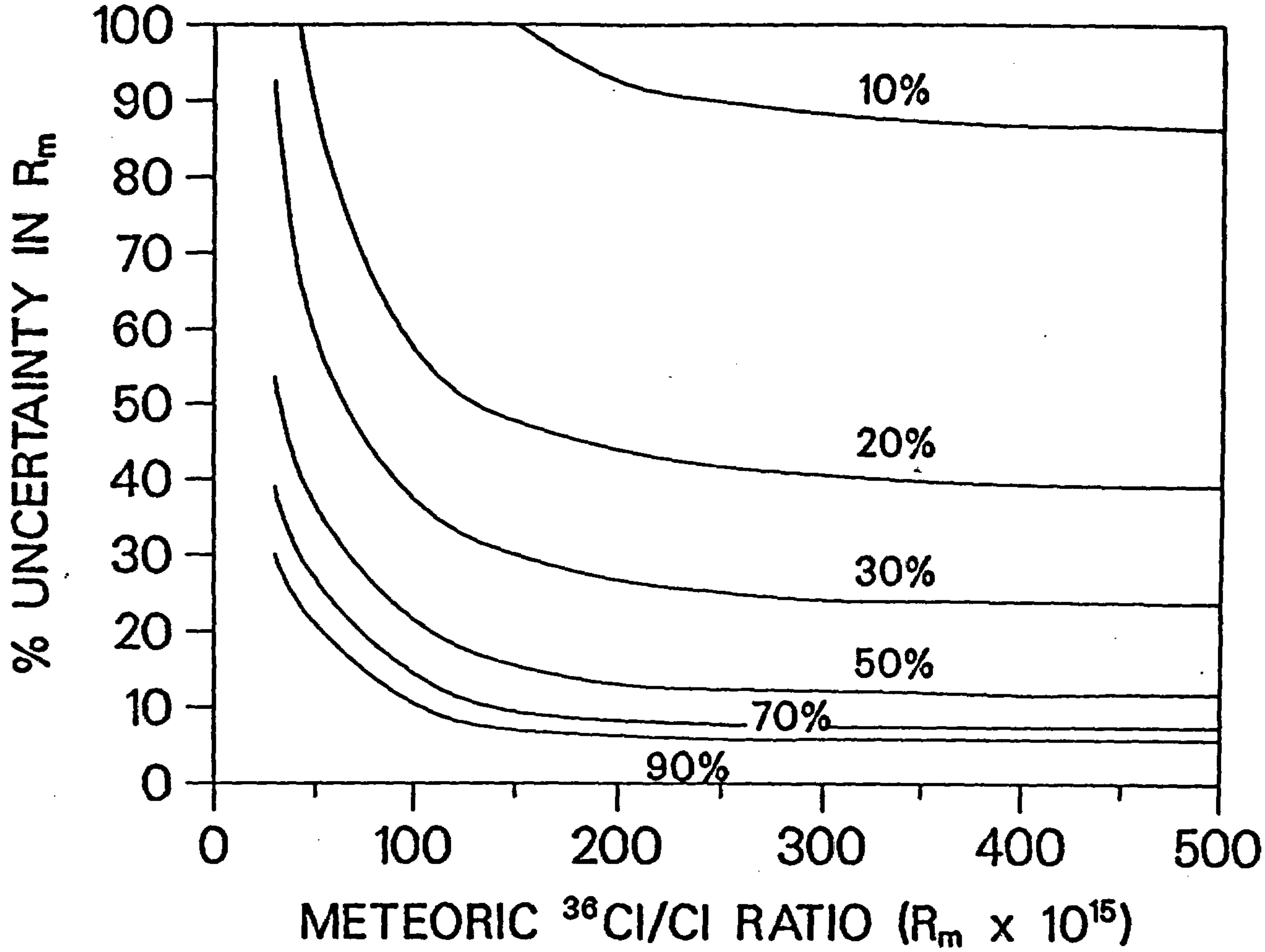

Figure 9. Relative uncertainty in the meteoric $^{36}Cl/Cl$ ratio as a function of the extent of dilution of the sample by rock chloride. Curves are labelled with the proportion of meteoric chloride, f_m, assumed to be present in the sample. Results plotted in this figure were calculated using equations (7) and (9b), assuming 5% analytical precisions for all Cl/Br ratios, a value of 2.0 (±0.4) x 10^{-14} for the rock $^{36}Cl/Cl$ ratio, and precisions for measured $^{36}Cl/Cl$ ratios taken from Figure 7.

$$\sigma_t = \left[\left(\frac{\partial t}{\partial R_m}\right)^2 \sigma^2_{R_m} + \left(\frac{\partial t}{\partial R_0}\right)^2 \sigma^2_{R_0} + \left(\frac{\partial t}{\partial R_r}\right)^2 \sigma^2_{R_r} \right]^{0.5} \qquad (10a)$$

$$= \left[\left(\frac{\sigma_{R_m}}{R_m}\right)^2 + \left(\frac{\sigma_{R_0}}{R_0}\right)^2 + \left(\frac{1}{R_0 - R_r} - \frac{1}{R_x - R_r}\right)^2 \sigma^2_{R_r} \right]^{0.5} \qquad (10b)$$

Figure 10 shows how the relative uncertainty in t varies as a function of f_m.

Uncertainties in the determination of water velocities. Results from Figure 10 can be used to estimate the uncertainty expected in average water velocity determinations for samples from the ESF. A depth of 325 m is assumed for the Main Test Level in the Topopah Spring welded unit Tsw2, and 425 m for the test level in the Calico Hills unit. Assuming that the relative uncertainty in the linear velocity will be the same as that for the residence time, Figures 11a and 11b shows how the uncertainty is expected to vary as a function of the velocity for a single sample from these levels. The uncertainty in the velocity estimate will decrease as the sample size n increases. From these figures, the practical limit for the ^{36}Cl tracer test appears to be linear velocities in the range of 0.4 to 5 mm/yr.

Lessons learned. Several lessons are learned from this statistical exercise. Clearly, the best results will be obtained by minimizing crushing of the rock samples prior to extraction of the soluble Cl. Analytical uncertainties for Cl/Br ratios on the order of 5% are expected. To ensure relative errors in f_m of 10% or less, then the crushing procedure should be such that $f_m > 0.7$. Even with high-quality Cl/Br analyses, rock flour from air-drilling operations will give rise to excessively low f_m values with unacceptably large uncertainties. The optimum sample form for rocks is as large chips or core.

Another lesson is the importance of establishing end-member ratios with minimal relative uncertainties, which will require that a sufficient number of appropriate samples be collected and analyzed. For determinations of meteoric Cl/Br samples, a large number of surface soil samples and soil profiles will be analyzed. For determinations of meteoric $^{36}Cl/Cl$ ratios, several soil profiles extending below the zone of bomb-pulse penetration are required. For determination of rock Cl/Br and $^{36}Cl/Cl$ ratios, several

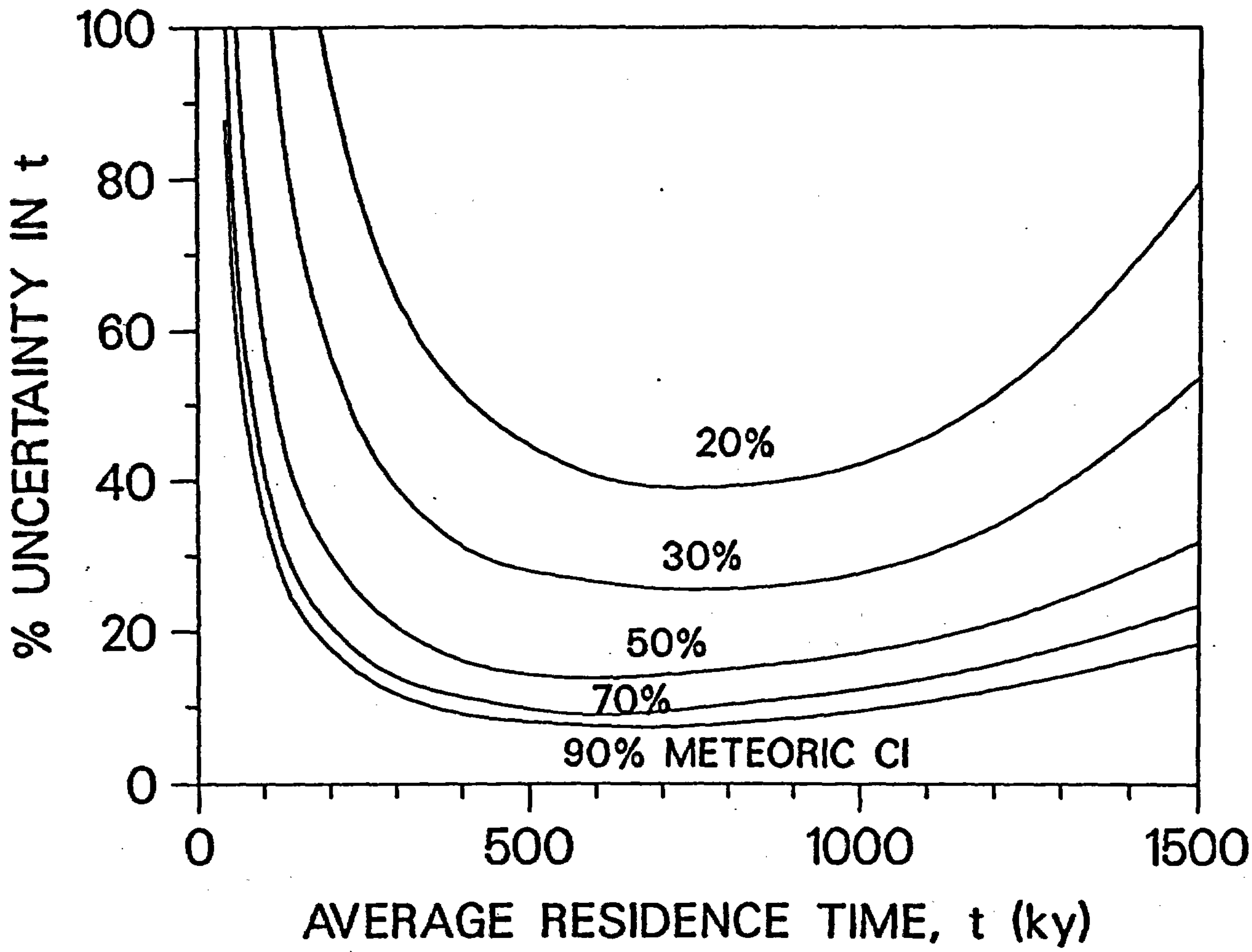

Figure 10. Relative uncertainty in the calculated residence time, t, as a function of the extent of dilution of the sample by rock chloride. Curves are labelled with the proportion of meteoric chloride, f_m, assumed to be present in the sample. Results plotted in this figure were calculated using equations (4) and (10b), under the same assumptions as those used to prepare Figure 9, and assuming a value of 5.2 (±0.5) x 10^{-13} for the initial meteoric $^{36}Cl/Cl$ ratio.

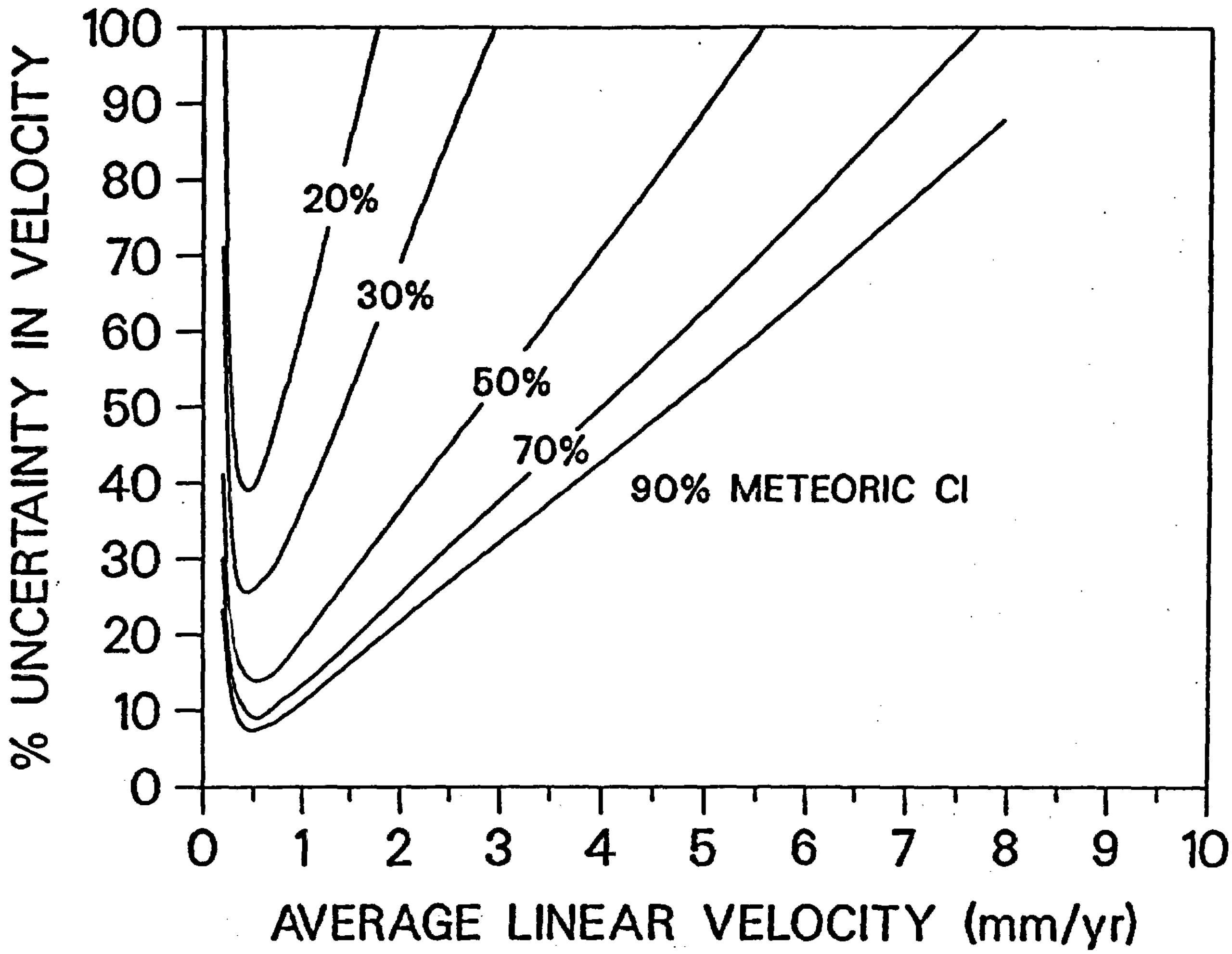

Figure 11a. Relative uncertainty in the average linear velocity determined for a sample collected at a depth of 325 m, corresponding to the average depth of the Main Test Level in the Topopah Spring welded unit, as a function of the extent of dilution of the sample by rock chloride. The uncertainty in the velocity estimate will decrease as the sample size n is increased.

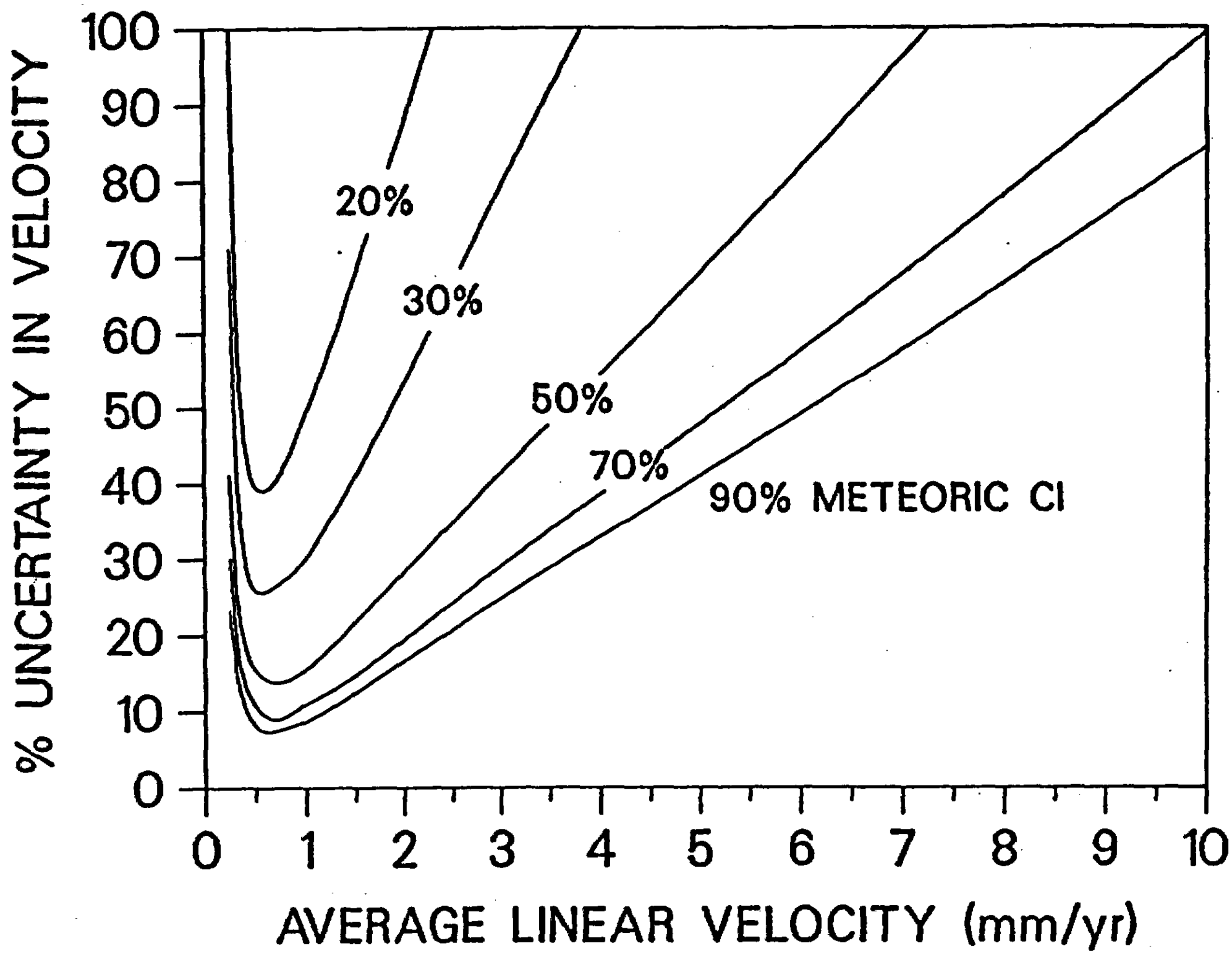

Figure 11b. Relative uncertainty in the average linear velocity determined for a sample collected at a depth of 425 m, corresponding to the average depth of the ESF Test Level in the Calico Hills unit, as a function of the extent of dilution of the sample by rock chloride. The uncertainty in the velocity estimate will decrease as the sample size n is increased.

samples of cores or large chips are needed of sufficient size to prepare several leachates by the step-leaching procedure (section 3.3.1).

Thirdly, a systematic evaluation of the error terms in each of the equations that comprise the ^{36}Cl age-dating model provides guidance for deciding where efforts should be directed in order to improve the precision of the age-dating measurements.

3.7 Representativeness of Velocity Determinations from the Exploratory Studies Facility

Data gathered from this study are expected to help describe infiltration processes within the repository block. Areal and vertical coverage of variables to be measured in the block by this study will be provided by the large number of surface-based vertical boreholes (Figure 4) which will be supplying samples to this study and by the current ESF design (Figure 6), which proposes ramp access and several kilometers of drifts in the Topopah Spring and Calico Hills units. A lack of obtainable or suitable sample material for analysis is not expected to be a problem. Possibilities for erroneous results or inconclusive interpretation of the data to be collected in this study is discussed in sections 2.2.5 (Potential for interferences from other field activities), and 3.4.3 (Limitations and assumptions for the interpretation of the ^{36}Cl data).

3.8 Quality Assurance Requirements

Activities in this study plan will be conducted in accordance with YMPO and LANL/YMP Quality Assurance requirements. Data from this study 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.

Technical procedures for the work in this study are shown in Table 4.

4.0 APPLICATION OF RESULTS

4.1 Resolution of Performance Issues

This study is important in that it will provide independent and unique information to address potential site suitability issues related to ground-water flow. The application of results in the site investigation work can be tied directly to resolution of key performance issues. The principal applications will be in assessments of ground-water fluxes and travel time through the unsaturated zone (Issues 1.1, 1.6, 1.8, and 1.9). The manner in which site information from this study will be used to resolve these issues is addressed in Section 1.0. Data from this study will support license application through testing and evaluation of unsaturated-zone flow and transport models that will be used by both site characterization and performance assessment. Chlorine-36 and other data derived from this study represent one approach to provide confidence in the application of these models to Yucca Mountain.

4.2 Interfaces with Other Site Characterization Studies

This study is part of Investigation 8.3.1.2.2, Description of the Unsaturated Zone Hydrologic System at the Site, which is directed at understanding the fundamentals of unsaturated flow and transport at Yucca Mountain. This study plan is coordinated with those in Investigation 8.3.1.2.2 through the sharing of common sampling locations (boreholes, trenches) among activities; the development of coordinated sampling plans for the ESF and surface-based drilling sites; and the sharing of data through the YMP Technical Data Base. All borehole and drillhole samples collected for this study will be obtained through an interface with the Sample Management Facility. Documentation for other samples collected for this study, such as soil profiles and surface soil samples, will be provided to the SMF following YMPO procedures. Information will be provided to, and obtained from, other studies primarily through the YMP Technical Data Base. Studies for which data from this study may be useful are discussed below.

Data from this study will principally be used in studies that comprise Investigation 8.3.1.2.2. Data generated by the Water Movement Test will provide information for determining the ground-water travel time in the unsaturated zone at Yucca Mountain and hence will be used to validate conceptual models of hydrologic flow in the unsaturated zone. Water movements at the site will be characterized through measurements of $^{36}Cl/Cl$ ratios as a function of depth below the surface. Information from ^{36}Cl data may be used for inferring rates of fracture flow relative to matrix flow. The vertical distribution of ^{36}Cl may permit assessment of the role of convective water movement relative to dispersive movement. (Limitations on the interpretation of ^{36}Cl data for these various applications have been discussed in sections 2.2.5 and 3.4.3.) Specific studies to which such data may be pertinent are listed below; the various tests involving ^{36}Cl analyses may provide an independent check on the results obtained by these studies. However, this study plan neither constrains nor is constrained by progress by these other studies and activities.

8.3.1.2.2.1 -	Characterization of Unsaturated-Zone Infiltration
8.3.1.2.2.3 -	Characterization of the Yucca Mountain Unsaturated Zone Percolation: Surface-Based Studies
8.3.1.2.2.4 -	Characterization of the Yucca Mountain Unsaturated Zone Percolation: Exploratory Shaft Facility Study
8.3.1.2.2.7 -	Hydrochemical Characterization of the Unsaturated Zone
8.3.1.2.2.8 -	Flow in Unsaturated Fractured Rock
8.3.1.2.2.9 -	Site Unsaturated-Zone Modeling, Synthesis and Integration

In addition, data from this study may also contribute to studies aimed at providing a description of the regional hydrologic system. The ^{36}Cl data obtained from the saturated zone may be useful for estimating zones of water mixing, potential sources of mixing end-members, and limits on water travel time. Such data may be applicable to Study 8.3.1.2.1.3 (Characterization of the Yucca Mountain Regional Ground-Water Flow System), 8.3.1.2.3.2 (Characterization of the Yucca Mountain Saturated-Zone Hydrochemistry), and 8.3.1.2.3.3 (Site Saturated-Zone Hydrologic System Synthesis and Modeling).

The use of rates of ^{36}Cl migration as an upper bound on ^{99}Tc 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, including, for example, Study 8.3.1.3.7.1 (Retardation Sensitivity Analysis). Ground-water travel time estimates from the Water Movement Test are also relevant to this latter study.

5.0 SCHEDULE AND MILESTONES

A tentative schedule for the work covered in this study is shown in Figure 12. The schedule information includes the sequencing, interrelations, and relative durations of the described activities, along with milestone numbers and titles from the master schedule presented in SCP Section 8.5.1.2. Interfaces among the studies and activities described in Section 4.2 are not shown because progress on the other studies neither constrain nor are constrained by progress on this study. The surface-based drilling program and ESF construction schedules are constraints on the progress of this study. Access to an accelerator mass spectrometer capable of analyzing ^{36}Cl with the required detection limit and sensitivity may also constrain progress.

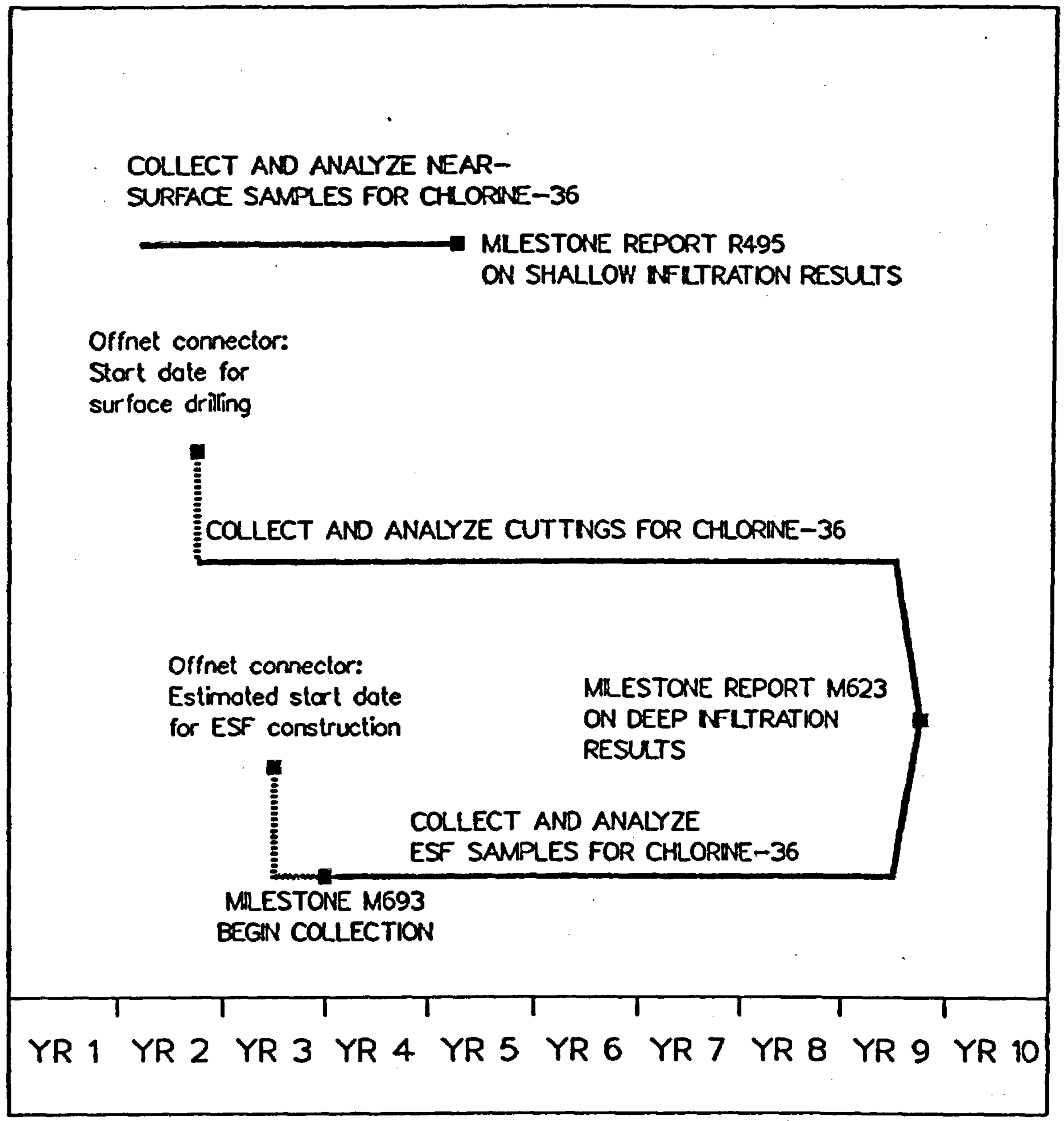

Figure 12. Anticipated progress in this study

6.0 REFERENCES

Andrews J.N., J-Ch. Fontes, J-L. Michelot, and D. Elmore, 1986. In-situ neutron flux, ^{36}Cl production and groundwater evolution in crystalline rocks at Stripa, Sweden, Earth and Planetary Science Letters, 77:49-58. (NNA.900824.0011)

Andrews J.N., S.N. Davis, J. Fabryka-Martin, J-Ch. Fontes, B.E. Lehmann, H.H. Loosli, J-L. Michelot, H. Moser, B. Smith, and M. Wolf, 1989. The in situ production of radioisotopes in rock matrices with particular reference to the Stripa granite, Geochimica et Cosmochimica Acta, 53:1803-1815. (NNA.910911.0061)

ASTM, 1990a. D1246-88, Standard Test Method for Bromide Ion in Water, Volume 11.01 Water and Environmental Technology, 1990 Annual Book of ASTM Standards, pp 293-295. (NNA.910911.0062)

ASTM, 1990b. D2216-80, Standard Method for Laboratory Determination of Water (Moisture) Content of Soil, Rock, and Soil-Aggregate Mixtures, Volume 04.08 Soil and Rock; Dimension Stone; Geosynthetics, 1990 Annual Book of ASTM Standards, pp 274-275. (NNA.910911.0063)

ASTM, 1990c. D4327-88, Standard Test Method for Anions in Water by Ion Chromatography, Volume 11.01 Water (I), 1990 Annual Book of ASTM Standards, pp 260-265. (NNA.910911.0064)

ASTM, 1990d. D4643-87, Standard Test Method for Determination of Water (Moisture) Content of Soil by the Microwave Oven Method, Volume 04.08 Soil and Rock; Dimension Stone; Geosynthetics, 1990 Annual Book of ASTM Standards, pp 859-862. (NNA.910911.0065)

ASTM, 1990e. D512-89, Standard Test Methods for Chloride Ion in Water, Volume 11.01 Water and Environmental Technology, 1990 Annual Book of ASTM Standards, pp 323-329. (NNA.910911.0066)

Bard E., B. Hamelin, R.G. Fairbanks, and A. Zindler, 1990. Calibration of the ^{14}C timescale over the past 30,000 years using mass spectrometric U-Th ages from Barbados corals, Nature, 345:405-410. (NNA.930119.0129)

Bentley, H. W., F. M. Phillips, and S. N. Davis, 1986a. "Chlorine-36 in the Terrestrial Environment," in Handbook of Environmental Isotope Geochemistry, J. C. Fontes and P. Fritz, Eds., Vol. IIB (Elsevier, Amsterdam), pp. 427-480. (NNA.910911.0067)

Bentley H.W., F.M. Phillips, S.N. Davis, M.A. Habermehl, P.L. Airey, G.E. Calf, D. Elmore, H.E. Gove, and T. Torgersen, 1986b. Chlorine-36 dating of very old ground water, 1. The Great Artesian Basin, Australia, Water Resources Research, 22:1991-2001. (NNA.910911.0068)

Beven K., and P. Germann, 1982. Macropores and water flow in soils, Water Resources Research, 18:1311-1325.

Bird J.R., N. Shahgholi, A. Jenkinson, A. Smith, L.K. Fifield, T. Ophel, and G. Allan, 1990. Problems of contamination in ^{36}Cl studies, Nuclear Instruments and Methods in Physics Research, B52:348-350. (NNA.910911.0069)

Blake G.R., 1965. Bulk density, pp 374-390. In C.A. Black (ed.), Methods of Soil Analysis. Part 1 Physical and Mineralogical Properties, Including Statistics of Measurement and Sampling, Number 9 in the series Agronomy (American Society of Agronomy, Inc., Madison, Wis.). (NNA.900208.0032)

Bloomsburg G., R.E. Williams, and J.L. Osiensky, 1989. Distribution of downward flux in unsaturated heterogeneous hydrogeology environments, Geological Society of America Bulletin, 101:1623-1630. (NNA.901228.0257)

Bowman R.S., and R.C. Rice, 1986. Transport of conservative tracers in the field under intermittent flood irrigation, Water Resources Research, 22:1531-1536.

Davis S.N., and E. Murphy, 1987. Dating Ground Water and the Evaluation of Repositories for Radioactive Waste. Washington D.C., U.S. Government Printing Office, Report NUREG/CR-4912, 181 p. (NNA.900522.0260)

Desaulniers, D. D., R. S. Kaufmann, J. A. Cherry, and H. W. Bentley, 1986. ^{37}Cl-^{35}Cl variations in a diffusion-controlled groundwater system, Geochimica et Cosmochimica Acta, 50:1757-1764. (NNA.900824.0009)

DOE (U. S. Department of Energy), 1984. 10 CFR Part 960, "Nuclear Waste Policy Act of 1982; General Guidelines for the Recommendation of Sites for the Nuclear Waste Repositories; Final Siting Guidelines," Federal Register, Vol. 49, No. 236 (Thursday, December 6, 1984), 47714-47770. (NNA.870506.0140)

DOE (U. S. Department of Energy), December 1988. "Site Characterization Plan, Yucca Mountain Site, Nevada Research and Development Area, Nevada," 8 vols., DOE/RW-0199, Office of Civilian Radioactive Waste Management, Washington, D.C. (HQO.881201.0002)

Eastoe C.J., Guilbert J.M., and R.S. Kaufmann, 1989. Preliminary evidence for fractionation of stable chlorine isotopes in ore-forming hydrothermal systems, Geology, 17:285-288. (NNA.910911.0070)

Elmore, D., P. W. Kubik, L. E. Tubbs, H. E. Gove, R. Teng, T. Hemmick, B. Chrunyk, and N. Conard, 1984a. The Rochester Tandem Accelerator Mass Spectrometry Program, Nuclear Instruments and Methods in Physics Research, B5:109-116. (NNA.900824.0010)

Elmore, D., N. Conard, P. W. Kubik, and J. Fabryka-Martin, 1984b. Computer controlled isotope ratio measurements and data analysis, Nuclear Instruments and Methods in Physics Research, B5:233-237. (NNA.900824.0012)

EPA (U. S. Environmental Protection Agency), 1985. 40 CFR Part 191, "Environmental Standards for the Management and Disposal of Spent Nuclear Fuel, High-Level, and Transuranic Radioactive Wastes; Final Rule," Federal Register, Vol. 50, No. 182 (Thursday, September 19, 1985), 38066-38089. (HQS.880517.0909)

Fabryka-Martin J., S.N. Davis, and D. Elmore, 1987. Applications of ^{129}I and ^{36}Cl in hydrology, Nucl. Instrum. Meth. Phys. Res., B29:361-371. (NNA.930119.0130)

Fabryka-Martin J., D.O. Whittemore, S.N. Davis, P.W. Kubik, and P. Sharma, 1991. Geochemistry of halogens in the Milk River aquifer, Alberta, Canada. Applied Geochemistry, 6:465-472. (NNA.910911.0097)

Feige Y., B.G. Oltman, and J. Kastner, 1968. Production rates of neutrons in soils due to natural radioactivity, Journal of Geophysical Research, 73:3135-3142. (NNA.910911.0071)

Fishman M.J., and L.C. Friedman, 1989a. Bromide, colorimetric, fluorescein, automated segmented flow, pp. 121-123 in Techniques of Water-Resources Investigations of the United States Geological Survey, Chapter A1 Methods for Determination of Inorganic Substances in Water and Fluvial Sediments, Book 5 Laboratory Analysis, U.S. Govt. Printing Office, Washington D.C.. (NNA.910911.0101)

Fishman M.J., and L.C. Friedman, 1989b. Chloride, colorimetric, ferric thiocyanate, pp. 149-150 in Techniques of Water-Resources Investigations of the United States Geological Survey, Chapter A1 Methods for Determination of Inorganic Substances in Water and Fluvial Sediments, Book 5 Laboratory Analysis, U.S. Govt. Printing Office, Washington D.C.. (NNA.910911.0098)

Fishman M.J., and L.C. Friedman, 1989c. Chloride, colorimetric, ferric thiocyanate, automated-segmented flow, pp. 151-153 in Techniques of Water-Resources Investigations of the United States Geological Survey, Chapter A1 Methods for Determination of Inorganic Substances in Water and Fluvial Sediments, Book 5 Laboratory Analysis, U.S. Govt. Printing Office, Washington D.C.. (NNA.910911.0099)

Fishman M.J., and L.C. Friedman, 1989d. Chloride, colorimetric, ferric thiocyanate, automated-discrete, pp. 155-156 in Techniques of Water-Resources Investigations of the United States Geological Survey, Chapter A1 Methods for Determination of Inorganic Substances in Water and Fluvial Sediments, Book 5 Laboratory Analysis, U.S. Govt. Printing Office, Washington D.C.. (NNA.910911.0100)

Fröhlich K., M. Ivanovich, M.J. Hendry, J.N. Andrews, S.N. Davis, R.J. Drimmie, J. Fabryka-Martin, T. Florkowski, P. Fritz, B. Lehmann, H.H. Loosli, and E. Nolte, 1991. Application of isotopic methods to dating of very old groundwaters: Milk River aquifer, Alberta, Canada, Appl. Geochem., 6:465-472. (NNA.930119.0131)

Gee G.W. and D. Hillel, 1988. Groundwater recharge in arid regions: review and critique of estimation methods, Hydrological Processes, 2:255-266. (NNA.910911.0072)

Heaton R., H. Lee, P. Skensved, and B.C. Robertson, 1990. Alpha-induced neutron activity in materials, Nuclear Geophysics, 4:499-510. (NNA.910911.0073)

James R.V., and J. Rubin, 1986. Transport of chloride ion in a water-unsaturated soil exhibiting anion exclusion. Soil Science Society of America Journal, 50:1142-1149. (NNA.910911.0074)

Johnston C.D., 1987. Distribution of environmental chloride in relation to subsurface hydrology, J. Hydrol., 94:67-88.

Kaufmann R.S., A. Long, H.W. Bentley, and S. Davis, 1984. Natural chlorine isotope variations, Nature, 309:338-340. (NNA.910911.0075)

Kaufmann R.S., A. Long, and D.J. Campbell, 1988. Chlorine isotope distribution in formation waters, Texas and Lousiana, American Association of Petroleum Geologists Bulletin, 72:839-844. (NNA.910911.0076)

Krupp H.K., J.W. Biggar, and D.R. Nielsen, 1972. Relative flow rates of salt and water in soil, Soil Science Society of America Proceedings, 36:412-417. (NNA.910911.0077)

Kubik P.W., P. Sharma, R.T.D. Teng, S. Tullai-Fitzpatrick, S. Datar, U. Fehn, and H.E. Gove, 1990. The AMS program at the University of Rochester, Nuclear Instruments and Methods in Physics Research, B52:238-242. (NNA.910911.0078)

Lehman L.L., 1992. Alternative conceptual model of ground water flow at Yucca Mountain, Proc., High Level Radioactive Waste Management Conf. (12-16 April 1992, Las Vegas NV), 1:310-320. (NNA.930119.0132)

Lehmann B.E., S.N. Davis, and J.T. Fabryka-Martin, 1993. Atmospheric and subsurface sources of stable and radioactive nuclides used for groundwater dating, Water Resources Research, in press. (NNA.930119.0133)

Lewis W.M., Jr., M.C. Grant, and J.F. Saunders, III, 1984. Chemical patterns of bulk atmospheric deposition in the State of Colorado, Water Resources Research, 20:1691-1704.

Lundström U., Å. Olin, and F. Nydahl, 1984. Determination of low levels of bromide in fresh water after chromatographic enrichment, Talanta, 31:45-48. (NNA.910911.0079)

Luten J.B., H.A. Das, and C.L. de Ligny, 1977. The determination of bromine and iodine in environmental water samples by thermal neutron activation and isotope exchange, Journal of Radioanalytical Chemistry, 35:147-155. (NNA.910911.0080)

Matthias A.D., H.M. Hassan, Y.-Q. Hu, J.E. Watson, and A.W. Warrick, 1986. Evapotranspiration estimates derived from subsoil salinity data, Journal of Hydrology, 85:209-223. (NNA.910911.0081)

Montazer, P., and W. E. Wilson, 1984. "Conceptual Hydrologic Model Flow in the Unsaturated Zone, Yucca Mountain, Nevada." Water Resources Investigations, US Geological Survey report USGS-WRI-84-4345, Denver, Colorado. (HQS.880517.1675)

Montazer P., E.P. Weeks, F. Thamir, S.N. Yard, and P.B. Hofrichter, 1985. Monitoring the vadose zone in fractured tuff, Yucca Mountain, Nevada, in Proceedings of the NWWA Conference on Characterization and Monitoring of the Vadose (Unsaturated) Zone. (National Water Well Association, Worthington, Ohio), pp. 439-469. (NNA.900924.0023)

Morrow C.M. and R.A. Minear, 1984. Determination of bromide in natural waters by ion chromatography using a concentrator column. Water Research, 18:1165-1168. (NNA.910911.0082)

Nitao J.J., T.A. Buscheck, and D.A. Chesnut, 1992. The implications of episodic nonequilibrium fracture-matrix flow on site suitability and total system performance. Proc., High Level Radioactive Waste Management Conf., (12-16 April 1992, Las Vegas NV), 1:279-296. (NNA.920225.0033)

Noble D.C., V.C. Smith, and L.C. Peck, 1967. Loss of halogens from crystallized and glassy silicic volcanic rocks, Geochim. Cosmochim. Acta, 31:215-223. (NNA.930119.0134)

Norris, A. E., K. Wolfsberg, S. K. Gifford, H. W. Bentley, and D. Elmore, 1987, Infiltration at Yucca Mountain, Nevada, traced by ^{36}Cl, Nuclear Instruments and Methods in Physics Research, B29:376-379. (NNI.880708.0062)

Norris A.E., H.W. Bentley, S. Cheng, P.W. Kubik, P. Sharma, and H.E. Gove, 1990. ^{36}Cl studies of water movements deep within unsaturated tuffs, Nuclear Instruments and Methods in Physics Research, B52:455-460. (NNA.910911.0083)

NRC (Nuclear Regulatory Commission), 1983. 10 CFR Part 60, "Disposal of High-Level Radioactive Wastes in Geologic Repositories Technical Criteria," Federal Register, Vol. 48, No. 120 (Tuesday, June 21, 1983), 28194-28229. (NNA.870406.0218)

Ogard A.E., J.L. Thompson, R.S. Rundberg, K. Wolfsberg, P.W. Kubik, D.Elmore, and H.W. Bentley, 1988. Migration of chlorine-36 and tritium from an underground nuclear test, Radiochimica Acta, 44/45:213-217. (NNA.910911.0084)

Ortiz T.S., F.B. Nimick, B.C. Whittet, and D.L. South, 1985. A three-dimensional model of reference thermal/mechanical and hydrological stratigraphy at Yucca Mountain, southern Nevada. Sandia National Laboratories, Report SAND84-1076, 71 pp.

Philbert F.J., 1982. Chapter 2, Major Ions, pp 67-129 in: Van Loon J.C. (ed.), Chemical Analysis of Inorganic Constituents of Water, CRC Press, Inc., Boca Raton, Florida. (NNA.910911.0085)

Phillips F.M., H.W. Bentley, S.N. Davis, D. Elmore, and G.B. Swanick, 1986a. Chlorine 36 dating of very old ground water, 2. Milk River Aquifer, Alberta, Canada, Water Resources Research, 22:2003-2016. (NNA.910911.0086)

Phillips F.M., B.D. Leavy, N.O. Jannik, D. Elmore, and P.W. Kubik, 1986b. The accumulation of cosmogenic chlorine-36 in rocks: a method for surface exposure dating, Science, 231:41-43.

Phillips F.M., J.L. Mattick, T.A. Duval, D. Elmore, and P.W. Kubik, 1988. Chlorine 36 and tritium from nuclear weapons fallout as tracers for long-term liquid and vapor movement in desert soils, Water Resources Research, 24:1877-1891. (NNA.910911.0087)

Phillips F.M., P. Sharma, and P. Wigand, 1991. Deciphering variations in cosmic radiation using cosmogenic ^{36}Cl in ancient rat urine, Amer. Geophy. Union Fall Meeting Program and Abstracts, p. 72. (NNA.930119.0135)

Roth K., W.A. Jury, H. Flühler, and W. Attinger, 1991. Transport of chloride through an unsaturated field soil, Water Resources Research, 27:2533-2541.

Scanlon B.R., 1991. Evaluation of moisture flux from chloride data in desert soils, J. Hydrol., 128:137-156. (NNA.930119.0136)

Scanlon B.R., 1992a. Evaluation of liquid and vapor water flow in desert soils based on Chlorine 36 and Tritium tracers and nonisothermal flow simulations, Water Resources Research, 28:285-297. (NNA.930119.0137)

Scanlon B.R., 1992b. Moisture and solute flux along preferred pathways characterized by fissured sediments in desert soils, J. Contaminant Hydrol., 10:19-46. (NNA.930119.0138)

Scanlon B.R., P.W. Kubik, P. Sharma, B.C. Richter, and H.E. Gove., 1990. Bomb chlorine-36 analysis in the characterization of unsaturated flow at a proposed radioactive waste disposal facility, Chihuahuan Desert, Texas, Nuclear Instruments and Methods in Physics Research, B52:489-492. (NNA.910911.0088)

Sharma M.L., and M.W. Hughes, 1985. Groundwater recharge estimation using chloride, deuterium and oxygen-18 profiles in the deep coastal sands of Western Australia, Journal of Hydrology, 81:93-109. (NNA.910911.0089)

Sharma P., P.W. Kubik, U. Fehn, H.E. Gove, K. Nishiizumi, and D. Elmore, 1990, Development of ^{36}Cl standards for AMS, Nuclear Instruments and Methods in Physics Research, B52:410-415. (NNA.910911.0090)

Sinnock S. (ed.), Y.T. Lin, and M.S. Tierney, 1986. Preliminary Estimates of Groundwater Travel Time and Radionuclide Transport at the Yucca Mountain Repository Site, SAND85-2701, Sandia National Laboratories, Albuquerque NM. (NNA.891129.0550)

Smith S.J., 1972. Relative rate of chloride movement in leaching of surface soils, Soil Science, 114:259-263. (NNA.910911.0091)

Synal H.-A., J.Beer, G. Bonani, M. Suter, and W. Wolfli, 1990. Atmospheric transport of bomb-produced ^{36}Cl, Nuclear Instruments and Methods in Physics Research, B52:483-488. (NNA.910911.0092)

Thomas G.W. and A.R. Swoboda, 1970. Anion exclusion effects on chloride movement in soils, Soil Science, 110:163-166. (NNA.910911.0093)

Vengosh A., A.R. Chivas, and M.T. McCulloch, 1989. Direct determination of boron and chlorine isotopic compositions in geological materials by negative thermal-ionization mass spectrometry. Chemical Geology (Isotope Geosciences Section), 79:333-343. (NNA.910911.0094)

Walker F.W., Parrington J.R., and F. Feiner, 1989. Nuclides and Isotopes (14th edn.), General Electric Company, San Jose, California, 57 p. (Readily available)

Walker G.R., I.D. Jolly, M.H. Stadter, F.W. Leaney, R.F. Davie, L.K. Fifield, T.R. Ophel, and J.R. Bird, 1991. Evaluation of the use of chlorine-36 in recharge studies. International Symposium on the Use of Isotope Techniques in Water Resources Development (Vienna, Austria, 11-15 March 1991). Paper No. IAEA-SM-319/2. (NNA.911127.0001)

Whitfield, M. S., 1985. "Vacuum Drilling of Unsaturated Tuffs at a Potential Radioactive-Waste Repository, Yucca Mountain, Nevada," in Proceedings of the NWWA Conference on Characterization and Monitoring of the Vadose (Unsaturated) Zone. (National Water Well Association, Worthington, Ohio), pp. 413-423. (HQS.880517.1869)

Whittemore D. O., 1988. Bromide as a Tracer in Ground-Water Studies: Geochemistry and Analytical Determination, Proceedings of the Ground Water Geochemistry Conference, held 16-18 February 1988 in Denver, Colorado, published by National Water Well Association, Dublin OH, pp 339-359. (NNA.910911.0095)

Wilson W.E., 1985. Unsaturated zone flux at Yucca Mountain, Nevada," attached to a letter from W.E. Wilson to D.L. Vieth dated 24 December 1985, U.S. Geological Survey, Denver CO, 10 pp. plus cover letter. (NNA.870323.0384)

Winchester J.W. and R.A. Duce, 1967. The global distribution of iodine, bromine, and chlorine in marine aerosols, Naturwissenschaften, 54:110-113. (NNA.910911.0096)

Wittwer C.S., G.S. Bodvarsson, M.P. Chornack, A.L. Flint, L.E. Flint, B.D. Lewis, R.W. Spengler, and C.A. Rautman, 1992. Design of a three-dimensional site-scale model for the unsaturated zone at Yucca Mountain, Nevada. Proc., High Level Radioactive Waste Management Conf. (12-16 April 1992, Las Vegas NV), 1:263-271. (NNA.930119.0139)

Yoshida M., K. Takahashi, N. Yonehara, T. Ozawa, and I. Iwasaki, 1971. The fluorine, chlorine, bromine, and iodine contents of volcanic rocks in Japan, Bull. Chem. Soc. Japan, 44:1844-1850. (NNA.930119.0140)

Young H.D., 1962. Statistical Treatment of Experimental Data, McGraw-Hill Book Company, Inc., New York, 172 p. (Readily available)

Zreda M.G., F.M. Phillips, D. Elmore, P.W. Kubik, P. Sharma, and R.I. Dorn, 1991. Cosmogenic chlorine-36 production rates in terrestrial rocks, Earth Planet. Sci. Lett., 105:94-109.

APPENDIX A

QUALITY ASSURANCE SUPPORT DOCUMENTATION

NOTE: The procedures listed in this Appendix may be superceded by updated revisions. A quality assurance grading report for the study's WBS element has been approved by the YMP Office in accordance with applicable YMP Office guidance.

TABLE A-1

APPLICABLE NQA-1 CRITERIA FOR SCP STUDY PLAN 8.3.1.3.6.2 AND HOW THEY WILL BE SATISFIED

NQA-1 Criteria	Documents Addressing These Requirements		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 organization is described in the LANL-YMP-QAPP.		
	TWS-QAS-QP-01.1	Interface Control Procedure	03/19/90
	LANL-YMP-QP-01.2	Stop Work Control	02/11/92
	LANL-YMP-QP-01.3	Conflict Resolution	02/11/92
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. The LANL QA program contains quality implementing procedures (QP) further defining the program requirements.		
	TWS-QAS-QP-02.3	Procedure for Readiness Review	03/19/90
	LANL-YMP-QP-02.4	Management Assessment	09/04/92
	LANL-YMP-QP-02.5	Selection of Personnel	09/30/91
	TWS-QAS-QP-02.7	Personnel Training	08/17/90
	LANL-YMP-QP-02.9	Personnel Proficiency Evaluations	08/20/92
	LANL-YMP-QP-02.11	Personnel Orientation	08/20/92
3. Design and Scientific Investigation Control	This study is a scientific investigation. The following QPs apply:		
	LANL-YMP-QP-03.5	Documenting Scientific Investigations	12/07/92
	TWS-QAS-QP-03.7	Procedure for Peer Review	05/24/89
	LANL-YMP-QP-03.17	Reviews of Software and Computational Data	01/25/91
	LANL-YMP-QP-03.18	Creation, Management, and Use of Computational Data	01/25/91

TABLE A-1 (continued)

NQA-1 Criteria	Documents Addressing These Requirements		Date of Issue
	LANL-YMP-QP-03.19	Documentation of Software and Computational Data	01/25/91
	LANL-YMP-QP-03.20	Software Configuration Management	01/25/91
	LANL-YMP-QP-03.21	Software Life Cycle	01/25/91
	LANL-YMP-QP-03.22	Verification and Validation of Software and Computational Data	01/25/91
	LANL-YMP-QP-03.23	Preparation and Review of Technical Information Products and Study Plan	03/16/92
	LANL-YMP-QP-03.24	Submittal of Design and Test-Related Information	06/15/92
	LANL-YMP-QP-03.25	Review of Design and Test-Related Information	06/15/92
4. Procurement Document Control	LANL-YMP-QP-04.4	Procurement of Commercial-Grade Items and Services	11/15/91
	LANL-YMP-QP-04.5	Procurement of Noncommercial-Grade Items and Services	12/23/91
5. Instructions, Procedures, Plans, and Drawings	Applicable parts of this criterion are covered in Item 6.		
6. Document Control	LANL-YMP-QP-06.1	Document Control	04/21/92
	LANL-YMP-QP-06.2	Preparation, Review, and Approval of Quality Administrative Procedures	09/30/91
	LANL-YMP-QP-06.3	Preparation, Review, and Approval of Detailed Technical Procedures	10/10/90
7. Control of Purchased Items and Services	Applicable parts of this criterion are covered in Item 4.		

TABLE A-1 (concluded)

NQA-1 Criteria	Document Addressing These Requirements		Date of Issue
8. Identification and Control of Samples and Data	LANL-YMP-QP-08.1	Identification and Control of Samples	02/28/92
	LANL-YMP-QP-08.3	Transfer of Data	07/10/92
9. Control of 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. Testing	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 Section 3 of the LANL-YMP-QAPP. The following QP also applies:		
	LANL-YMP-QP-12.1	Control of Measuring and Test Equipment	06/05/92
13. Handling, Shipping, and Storage	TWS-QAS-QP-13.1	Procedure for Handling, Storage, and Shipping Equipment	11/03/89
14. Inspection, Test, and Operating Status of Engineered Items	This criterion has been determined to be inapplicable to the scope of work of the LANL YMP.		
15. Control of Nonconformances	TWS-QAS-QP-15.2	Deficiency Reporting	04/03/90
16. Corrective Action	LANL-YMP-QP-16.2	Trending	09/04/92
	LANL-YMP-QP-16.3	Deficiency Reports	03/23/92
17. Records	LANL-YMP-QP-17.4	Records Preparation	02/28/92
	LANL-YMP-QP-17.5	Records Processing	02/28/92
18. Audits	LANL-YMP-QP-18.1	Audits	03/07/91
	TWS-QAS-QP-18.2	Surveys	05/30/90
	TWS-QAS-QP-18.3	Auditor Qualification and Certification	05/30/90

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